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PRESENT STATUS OF LITHIUM DIFFUSED SILICON SOLAR CELLS

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**Present Status of Lithium-
Diffused Silicon Solar
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Greenbelt, Maryland**

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PRESENT STATUS OF LITHIUM-DIFFUSED SILICON SOLAR CELLS

I. INTRODUCTION

The present report is concerned with lithium-diffused silicon solar cells.

First, a brief history of work on lithium-diffused semiconducting silicon will be given, and a few experimental results on the radiation effects and the after-effects will be discussed. Second, the radiation damage and recovery of lithium-diffused solar cells will be analyzed from the physical point of view. Finally, experimental results of the initial efficiency, and the subsequent radiation damage recovery will be discussed.

The solar cells used in this study were made by RCA Laboratories, Heliotek, Hoffman Electronics Corp., and Texas Instruments, Inc. The enthusiasm and active cooperation of these suppliers are appreciated. The experiments were performed by Messrs. G. Meszaros, Y. M. Liu, and W. McGunigal. The discussions among the members of the Radiation Physics Section of Goddard Space Flight Center were of great help in planning and carrying out the program. In addition, encouragement of Mr. M. Schach on this project is hereby acknowledged.

II. HISTORICAL REVIEW

Diffusion of lithium (Li) in silicon (Si) was first investigated by Fuller and Ditzenberger (Reference 1) by the p-n junction method, in which the location of the junction, which is formed by diffusing Li into p type Si, is measured.

They found that

$$D = 94 \times 10^{-3} \exp\left(-9.111 \times \frac{10^3}{T}\right) \text{ cm}^2 \text{ sec}^{-1}, \quad (\text{II} - 1)$$

D is the diffusion coefficient and T is the absolute temperature. This leads to a room-temperature value of $D = 1.3 \times 10^{-14} \text{ cm}^2 \text{ sec}^{-1}$. At 400°C, D will be $3.58 \times 10^{-9} \text{ cm}^2 \text{ sec}^{-1}$.

In a later work (Reference 2), Fuller and Severiens gave, by measuring mobility vs applied field,

$$D = 2.3 \times 10^{-3} \exp\left(-7.65 \times \frac{10^3}{T}\right) \text{ cm}^2 \text{ sec}^{-1}. \quad (\text{II} - 2)$$

This would give $D = 2.0 \times 10^{-14} \text{ cm}^2 \text{ sec}^{-1}$ at room temperature and $2.5 \times 10^{-8} \text{ cm}^2 \text{ sec}^{-1}$ at 400°C. In this experiment, Li is again diffused into p-type Si. Because there is an interaction and precipitation of Li in p-type, Boron (B) doped Si (see Reference 3), the value of D might be different from that obtained when Li is diffused into n-type Si or pure Si, but no data pertaining to these cases are available. Furthermore, since Li interacts effectively with oxygen (O) to form LiO^+ or other forms of the oxide, which are stable at room temperature (Reference 4), it should be noted that all these diffusion studies were carried out with Si crystals grown from a quartz crucible, i. e., Si of high O content. A recent work of Gilmer, et al. (Reference 5) gives an up-to-date and complete study of Li-O complexes as donor impurities in Si.

The investigation of the existence of the interaction of Li with the complex defects introduced by irradiation was initiated by Vavilov and his co-workers (Summarized in Reference 6). By observing the change of the characteristics of the temperature dependence of carrier concentration, and of the lifetime of minority carriers in p-type Si, with or without Li doping, the following observations were made:

1. In n-type Si which was obtained by inversion from p-type Si, radiation damage results in reversion to the original p-type. This is explained as due to the interaction of Li with the radiation-induced defect. The donor concentration contributed by Li is reduced. This effect is called precipitation by Vavilov.

2. The rate of production of A-centers (O vacancy complex) in Si containing Li is less than that in Si containing phosphorous (P) or arsenic (As). This is attributed to the formation of the LiO^+ complex. Thus, the effective oxygen concentration which provides a component of the A-center is reduced.

3. The degradation of the lifetime due to electron irradiation in Si containing Li shows a saturation for 1 Mev electrons after about $1 \times 10^{15} \text{ e/cm}^2$. In Si containing P but no Li, no such saturation, indeed, only a continuous degradation, is observed.

We will discuss more specifically the first two aspects later. The third aspect has not been verified (Reference 7). Although it was not expressly stated, presumably all crystals studied by Vavilov et al. have a high oxygen content.

Vavilov attributes the idea that the disappearance of defect centers is due to the interaction of impurity centers with structural defects, to Roberts et al. (Reference 8). In their brief paper, Roberts et al. found an increase in the majority carrier lifetime with an increase of the oxygen concentration, in contradiction to the usual expectation that any departure from crystal perfection will reduce the lifetime. These authors presented an interesting hypothesis that there is an unknown center which either can be nullified by chemical interaction with oxygen or competes with oxygen for a place in the lattice. This point has not been

further verified. In the meantime, a successful application of this idea to the case of defects introduced by irradiation is clearly shown by Vavilov et al.

It was the lecture of Vavilov (Reference 6) that revived the interest of Rappaport in investigating the introduction of Li into Si solar cells. * Immediately a study was made of the surface barrier diode with Li diffusion (Reference 9). The degradation of the diffusion length was measured as a function of the radiation flux. A recovery of the diffusion length was observed after cessation of the radiation and a waiting period of several hours.

An attempt was made to apply this idea to solar cells with the following configurations: (1) n-on-p solar cells, (2) p-on-n solar cells made from pulled crystals, and (3) p-on-n solar cells made from float zone crystals but with a high P content ($\sim 1 \Omega \text{ cm}$ or less). In all cases, results the same as those observed in the case of the surface barrier diode were not obtained.

It was the contribution of Wysocki, at this point, to experiment on p-on-n solar cells made from float zone crystals with a low P content (larger than 10 cm). The result was spectacular: For the first time, a practically complete recovery at room temperature was observed in the solar-cell performance which had been degraded by the radiation. For 1 Mev electron irradiation and for an accumulated flux of $1 \times 10^{14} \text{ e/cm}^2$, a complete recovery was observed in a matter of hours. If the flux is higher than this value, recovery is less complete. This phenomenon is also dependent on the rate of radiation flux. The above irradiation was carried out at a rate of 1×10^{14} to $1 \times 10^{16} \text{ e/cm}^2\text{hr}$. If the flux rate is $5 \times 10^{12} \text{ e/cm}^2\text{hr}$ in fact, because the rate of the recovery is compatible with the rate of damage introduction, no damage is observed at all. All these observations have an analogy in the annealing of n-on-p solar cells (Reference 10). The critical flux levels are equal in both cases, within the accuracy of the experiment. If recovery is permitted intermediately by reducing the radiation rate,

*Private communication.

the recoveries are complete in both cases. In fact, in this way the total flux can exceed the critical flux obtained the result of a fast irradiation rate.

This brings the status up to early 1966. The progress made since is the main material of the present report.

III. PHYSICAL MODELS OF SPONTANEOUS ANNEALING

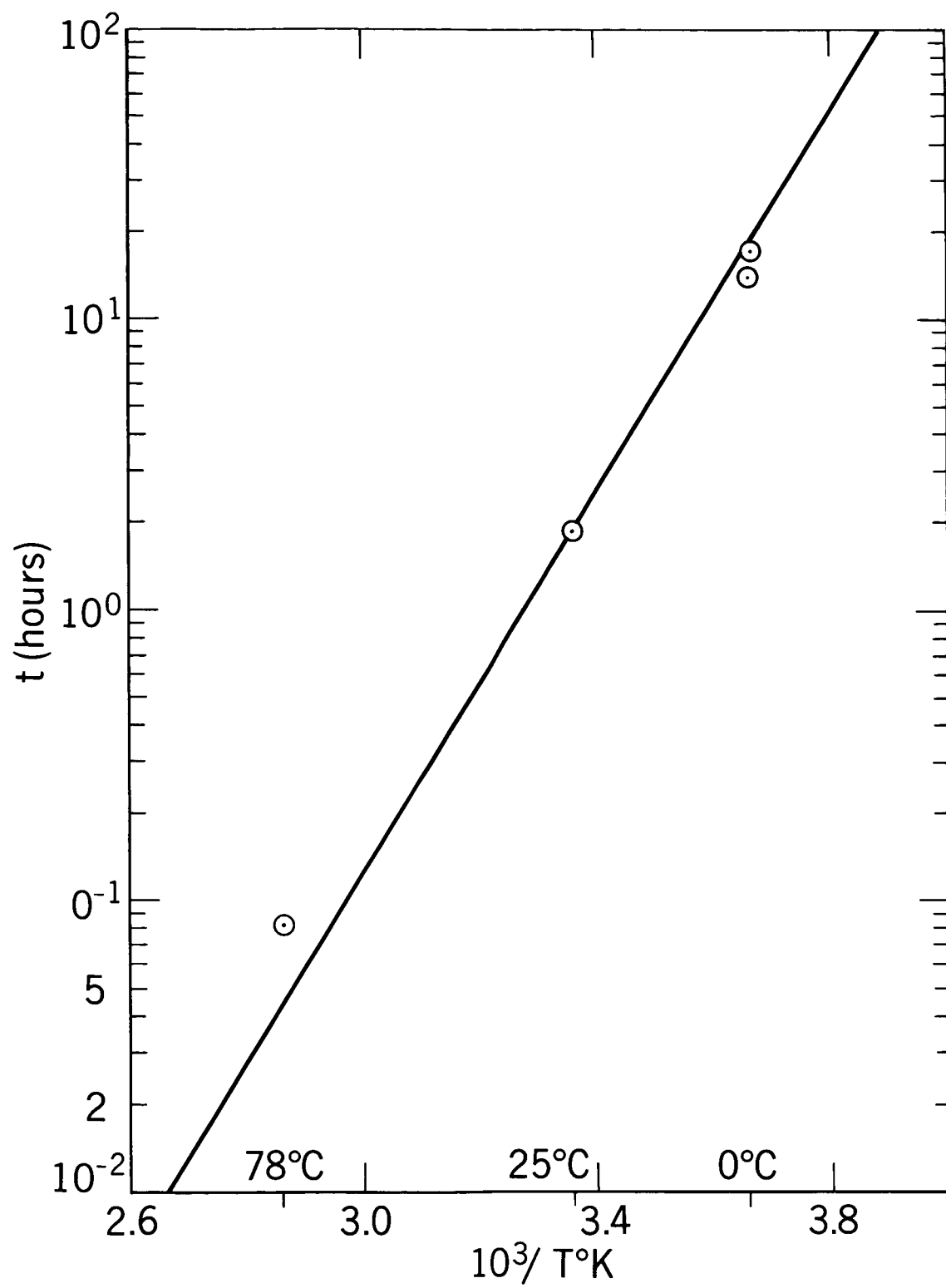
The present models of the mechanism of the spontaneous annealing of Li-diffused solar cells are based on indirect observations and are mostly speculative. Two different models exist, one in which the role of Li is to compensate the defects (Reference 11) and one in which the role of Li is to form metastable defects (Reference 12).

The Role of Li is to Compensate the Defects

When the Li is to compensate the defects, Li^+ is moving randomly in the crystal lattices by diffusion. When a defect due to the radiation occurs near the instantaneous site of Li^+ , a defect representing a recombination or trapping state to the minority carriers in the n-region would have a negative charge; therefore, an interaction with Li^+ would lead to a neutralization of the charge state and a consequent annealing.

One support of this model is the experimental data of the temperature dependence of the recovery time (Fig. III-1). The isothermal recovery times τ at 0° , 25° , and 78°C are measured at room temperature. For this limited temperature range and radiation fluxes of 1 and $3 \times 10^{14} \text{ e/cm}^2$ of 1 Mev electron radiation the plot of τ vs. $1/T$, where T is the absolute temperature, is a straight line whose slope of 0.66 ev corresponds closely to the value of the activation energy of diffusion previously reported (Reference 1 and 2). But in view of the lack of the precise value of the activation energy of diffusion and the limited temperature data used, this cannot be treated as conclusive evidence. There is also some evidence that, even if the annealing is controlled by the diffusion of Li to the compensation site, a complexity could result from the recent observation that the recovery rate is dependent on the density of the defect.* This is not taken into account in the treatment of the diffusion theory from which the experimental results were analyzed.

*P. H. Fang and Y. M. Liu (to be published).



III-1

Probably the strongest argument against this model is that the center which is assumed to be annealed by Li cannot be identified with any presently known defect.* As described in the beginning, the spontaneous annealing is observed in float zone grown, but not pulled, Si; this excludes the importance of the A-center. Secondly, the annealing is observed in moderate-to-low P concentration material in, and low concentration B material which undergoes subsequent conversion to n-type Si; thus the recovery is independent of E-center defects. Finally, the divacancy defect cannot be important because of its low rate of production.

Another difficulty of this model is the following observation: Solar cells, after electron irradiation and after an almost complete recovery, are heat-treated up to 350°C for several hours to look for signs of reverse annealing. In this temperature range, all known defects are annealed by a dissociation of the vacancy from the complex.* In the case of Li, therefore, a dissociation of Li from the defect, or a dissociation of the defect itself, should lead to an observation of a change in photovoltaic characteristics. The result is negative — no ascertainable changes were observed.

This deductive reasoning suggests the simpler and more direct model which will be discussed next.

The Role of Li is to Form Metastable Defects

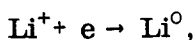
When high-energy particles enter the Si lattice, vacancy-interstitial pairs will be created. Both vacancies and interstitials are highly unstable at room temperature, but vacancies are less unstable and most of them are transformed into stable states by forming complexes with impurities or with other vacancies. In float zone Si with Li as the major impurity, there is a great probability that there will be an interaction between the vacancy and Li. This interaction has been observed by Vavilov (Reference (6)). Goldstein has conjectured the existence of a Li-V (where V denotes vacancy) complex through electron paramagnetic resonance studies (Reference 13).

*G. Watkins, loc. c.f. Reference 6, p. 97.

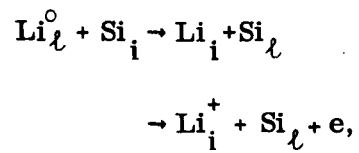
The model of the Li-V complex is an interstitial Li^+ and a V^- in a lattice position (Reference 12). Since Li^+ forms a stable state at an interstitial position in the regular Si lattice, even under Coulomb attraction by one of the four nearest neighbor Si ions, the Li^+ probably could maintain a metastable state. The annealing mechanism in this model, the activation energy of the recovery time in the microscopic diffusion, is assured to be approximately equal to that of the macroscopic diffusion.

Much basic information is needed to determine the more valid of the present phenomenological models. This knowledge is equally needed in the evaluation and the projection of the present result to technical applications. In this respect, an important question is that of stability over extensive periods of time and over a wide range of temperature. Also a very interesting question is whether the recoverability from a large dosage of radiation flux is limited by the initial Li concentration. If each annealing requires one Li^+ ion in the crystal, eventually, when the number of vacancies becomes approximately equal to the number of Li^+ ions, the spontaneous recovery will have ceased. As will be discussed later, we have noted that the defect production rate as well as the degree of recovery depends appreciably on the concentration of the instantaneous defect density, and, under some dynamic conditions, it seems that the extent of recovery after an extensive accumulated flux dose does not seem to be limited by the available Li concentration. A possible mechanism will now be given.

According to the second model of annealing, the recovery of the radiation damage is due to the occupancy of the lattice position by Li^+ of the Li^+-V^- complex defect. This position requires the trapping of an electron by Li^+ , thus,



is thermodynamically not stable. The most stable configuration is



which describes an interchange between interstitial (i) and lattice (l) positions and a subsequent capture of a hole. Two sources of Si_i are

1. A Frankel defect of thermal origin, and
2. In the case of simultaneous irradiation, continuously created Si_i .

The existence of such processes will be vital in determining the ultimate limitation of the practical use of Li solar cells.

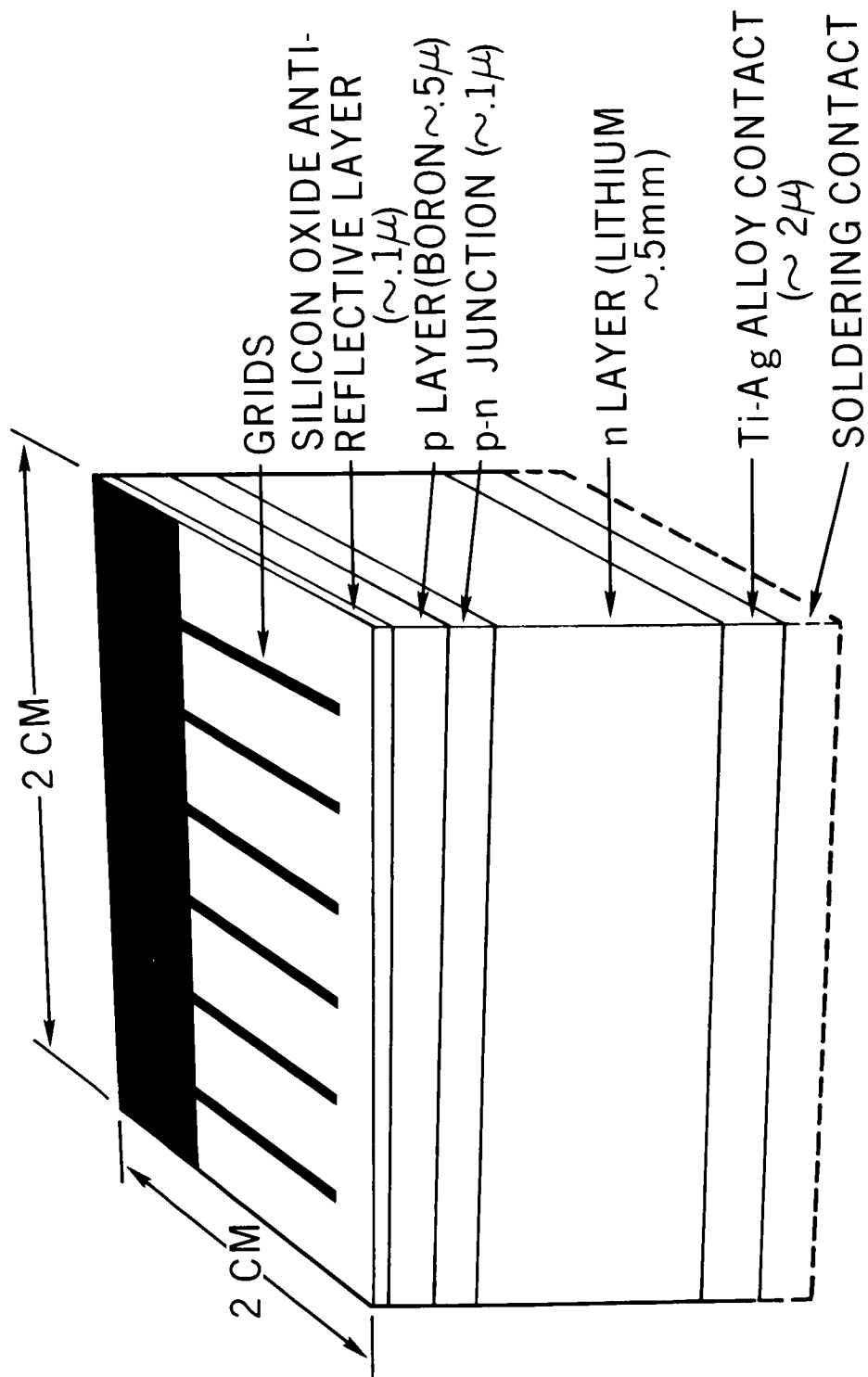
IV. DIFFERENT LI SOLAR CELLS

On the basis of the present knowledge of the interesting properties of the interaction of the radiation-induced defect with Li, four types of Li solar cells have been developed:

- Type I. p-on-n with float zone Si,
- Type II. p-on-n (phosphorus) on n (lithium) with float zone Si,
- Type III. p-on-n with pulled Si, and
- Type IV. n-on-p with float zone Si, with a relatively small Li concentration in the p-region.

Type I Cell. The Type I cell is shown in Figure IV-1. This is the principal type of Li cell. The important constituent of the cell is a Si crystal of low O content, a p (up to now, boron)-type front region, and Li as the dominant donor in the n-region. Most discussions up to now were based on this type of solar cell, and it will be described exclusively in later sections. Since a sufficiently specific description of the cell without misnomer requires a cumbersome expression and since the cell will be frequently referred to in the future, it seems most desirable to coin a special name for this type of cell. It is remarkable that the cells were made by Wysocki before we knew or had any clear physical picture of the nature of the radiation damage. The foresight of Wysocki (Reference 7) was instrumental in the cells discovery and it seems only proper to designate this type of cell as the Wysocki cell. Therefore, the type I cell will be so designated hereafter.

Type II Cell. The principle of the type II cell is based on the following observation: Because of a compensation near the p-n junction and because of the diffusion profile configuration, there is a depletion of Li concentration in the n-region near the p-n junction. With irradiation, further reduction of free Li results, because of a complex formation between Li and the radiation-induced



V-1

defect. This diminishing of Li will be most effective near the junction because of the original comparatively lower Li content in this region.

An approach to solving this problem is to make double n-layers*. The first n-layer, consisting of the p-n junction and the immediate n-region, is a few microns thick and is made by the diffusion of P into Si, and the layer thickness is controlled either by a diffusion or by a subsequent lapping. The p-layer is then diffused into the phosphorus n-layer to form the p-n junction. A Li diffusion, in the same manner as in the Wysocki cell, is then performed.

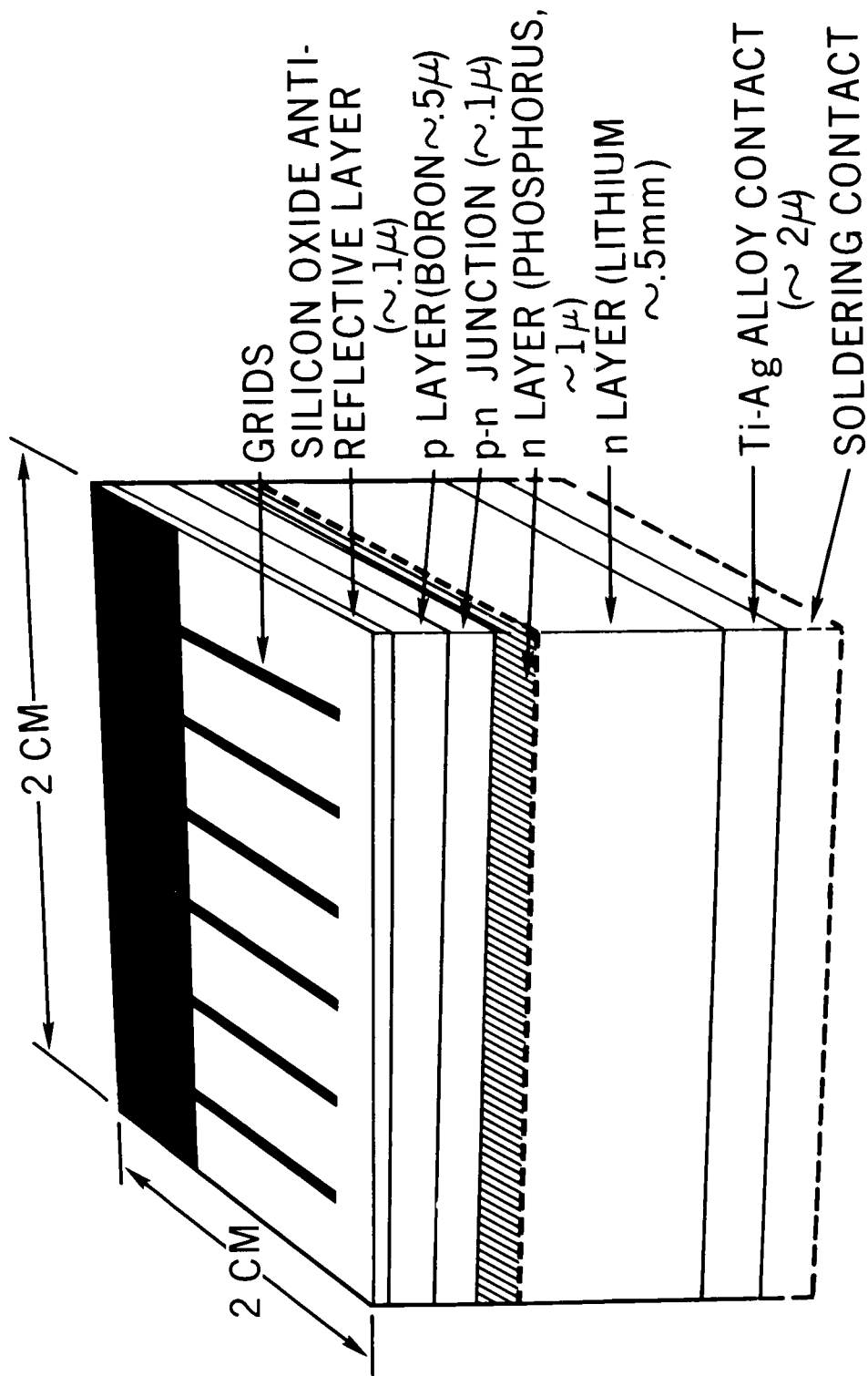
Figure IV-2 is a diagram of a Li Type II cell. The radiation damage property of the type II solar cells will be discussed in the Section entitled "Some Experimental Results Pertaining to Space Applications."

Type III Cell When Si of high O content, as is found in pulled crystal, is used, the defect induced by the radiation is stable at room temperature. However, an effective annealing stage occurs at a temperature of 120° to 150° C (Figure IV-3.) This is to be compared with the effective annealing temperature of about 400° C in the usual n-on-p solar cell.† The nature of this defect has been conjectured as a vacancy forming a complex with the Li-O pair.

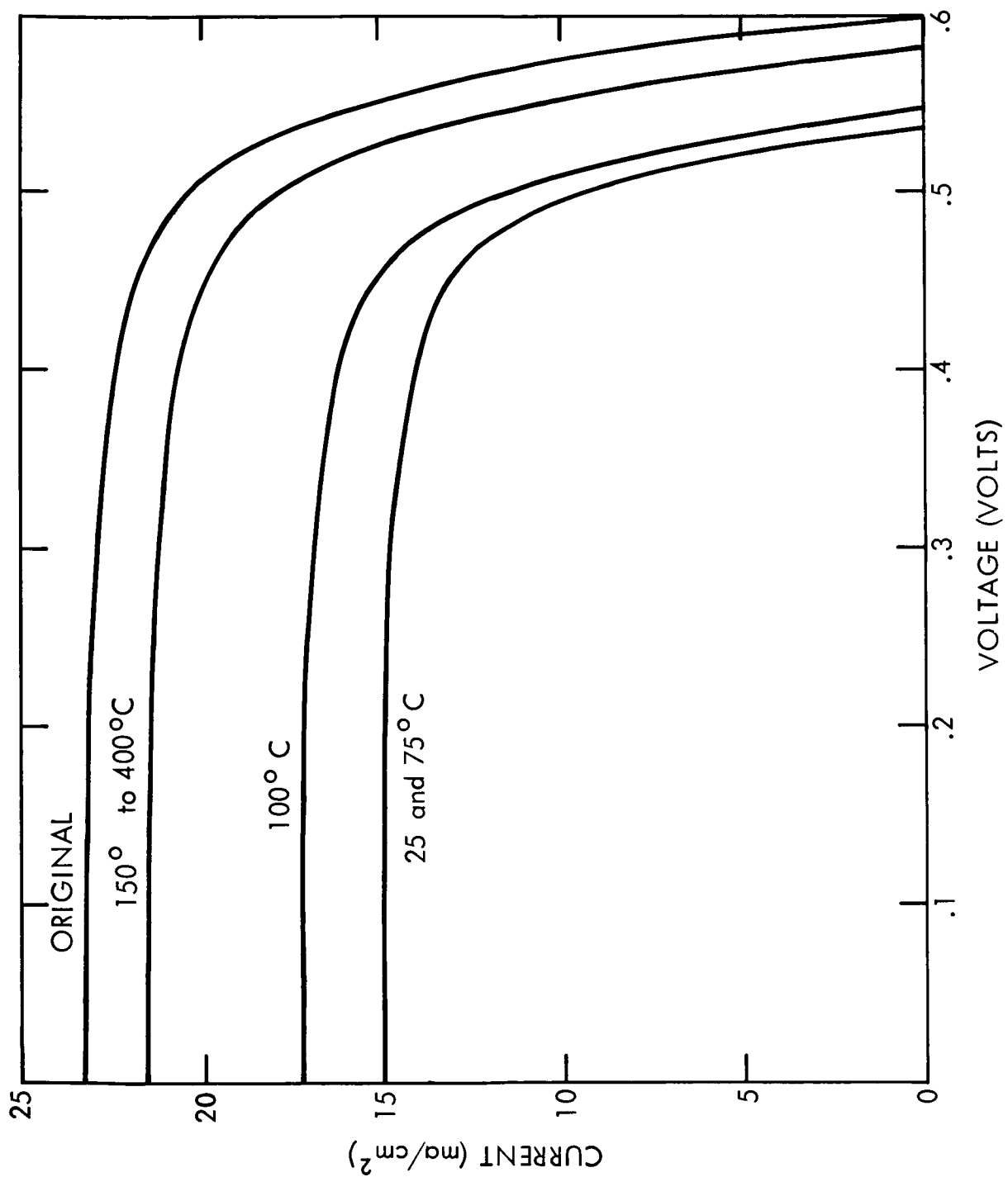
From a practical point of view, if a high-temperature annealing is to be used as a means of solving the problem of the radiation damage, a 150° C annealing is much to be preferred from the point of view of both the demand of the stability of the components and connections in the annealing machinery and in the required thermal power of annealing.

*P. H. Fang, NASA Goddard Space Flight Center (Patent pending).

†P. H. Fang, Astron. Acta (to appear).



V-2



V-3

Type IV Cell. In the original work of Vavilov (Reference 6) an interesting observation was reported: In p-type (B-doped) Si (Reference 3), when Li is introduced, an over-compensation resulted in the conversion to n-type Si. A subsequent electron irradiation showed a reversion to p-type. The interpretation was that Li-B pairs are formed and the effective original acceptors are reduced in number. With electron irradiation, the defects formed (presumably single vacancies) interact strongly with Li, B is reliberated, and the effect results in a restoration of the majority carrier concentration.

Suppose now a n-on-p solar cell is made from p-type Si with a high concentration of B, say, corresponding to 1 Ω -cm. If the introduction of Li resulted in the formation of p-type Si of 10 Ω -cm, the solar cell is now a 10 Ω -cm n-on-p cell. After irradiation of this type of cell, assuming there is no radiation effect other than the reversion of high resistivity p-type to low resistivity p-type, a radiation-induced improvement in the solar-cell performance could be expected.

V. LI SOLAR CELL TECHNOLOGY

The preparation of Li solar cells has been described previously in the work of Topfer et al (Reference 14). More up-to-date reports on the technological processes will become available in the near future (Reference 15). Therefore, the description here will emphasize the physical aspects and will leave out the technical processing.

Figure IV-1 shows the schematic of a p/n, Li-diffused, Si solar cell. In the fabrication, the only difference from the ordinary p/n solar cell of B (p-type) and P (n-type), is that a Li diffusion is carried out from the base of the solar cell. Since the diffusion coefficient of Li is enormously greater than that of the presently used p-layer dopant (B), the Li diffusion is carried out after the high temperature process of p-n junction formation by B diffusion. The Li diffusion is then carried out near 400°C.

There are three commonly used methods of diffusing Li into Si:

1. Paint the Si with a Li-oil emulsion. This is the simplest method, but the amount of Li applied to the surface cannot be easily controlled. The diffusion is carried out in a high-temperature furnace.
2. Immerse the Si in a Li-Sn alloy bath. The temperature of the alloy in the liquid state is about 400°C. The diffusion time required is usually longer than in 1, but a better uniformity can result. This method also presents some difficulty in the protection of the p-layer and preventing edge leakage.
3. Evaporate Li into Si with a subsequent diffusion. This method allows for better control of the amount of Li applied to the Si surface. In comparison with the Li-oil emulsion, since the oil is much less pure, the Li evaporation directly from Li metal reduces the introduction of unknown or undesired impurities.

On the problem of the solubility of Li in Si, the following parameters are probably important and are presently largely unknown.

1. The optimum concentration of Li. From the radiation damage and annealing point of view, high Li content is desirable. But high Li content could also result in a larger carrier removal rate, as observed in an analogous case in p-type Si (Reference 16).

According to the experimental data of Pell (Reference 17), the solubility of Li at 27°C is 10^{13} cm^{-3} . The concentration of Li obtained by the current diffusion practice is of the order of 10^{16} to 10^{17} cm^{-3} . Therefore, the system is highly super-saturated. The consequence is an expectation of the precipitation. The quantitative value of the precipitation time is unknown at the present.

2. The effect of the impurity on the solubility of Li. There is an obvious difference in the case of p-type Si from that of n-type Si. This is qualitatively explained by the electrolytic solution model (Reference 18). The Si specimens used in earlier experiments were pulled crystal of impurity concentration, before the Li diffusion, of the order of 10^{14} to 10^{16} impurities cm^{-3} . Since in the pulled crystal the O content is greater than 10^{16} cm^{-3} and there is a strong interaction between Li and O to form LiO^- and Li_2O , the problem of what will be the solubility of Li in float zone Si where the O content will be two or more orders of magnitude less than that in pulled crystal is still to be investigated.

Another problem is the solubility of Li in very pure Si of impurity content of the order of 10^{13} cm^{-3} , corresponding to a resistivity of about $10^3\Omega\text{-cm}$. If one is to emphasize the room temperature annealing, any impurity, other than Li, should be minimized because, according to present knowledge, the defects associated with all impurities, except those of Li, are stable up to 150°C or higher. Therefore, it is natural to start with Si material with as low an impurity concentration as possible.

3. The effect of dislocations on the solubility of Li. Two kinds of crystals used currently in Li solar cells are float zone and Lopex.* While float zone material has a dislocation concentration in the range of 10^{14}cm^{-3} , Lopex has about two orders of magnitude less. There is some indication that the diffusion coefficient of Li in Lopex material is smaller than in float zone material.**

4. In connection with the solubility, there is also an interesting problem of the concentration profile. Since in the Li solar cell the donor is introduced by diffusion, and, furthermore, since a p-n junction is present during this diffusion, the region having an excessively high concentration, greater than 10^{19}cm^{-3} , of acceptors behaves as an infinite sink. Therefore, the concentration of Li cannot approach a constant value throughout the depth of the cell. This unintentional concentration profile actually satisfies the configuration of a p-on-n drift field solar cell and was considered by Ross long before its radiation and annealing properties were investigated.***

*A product of Texas Instruments, Inc.

**D. Kendall (private communication).

***B. Ross (assigned to Hoffman Electronics Corp.). Patent pending.

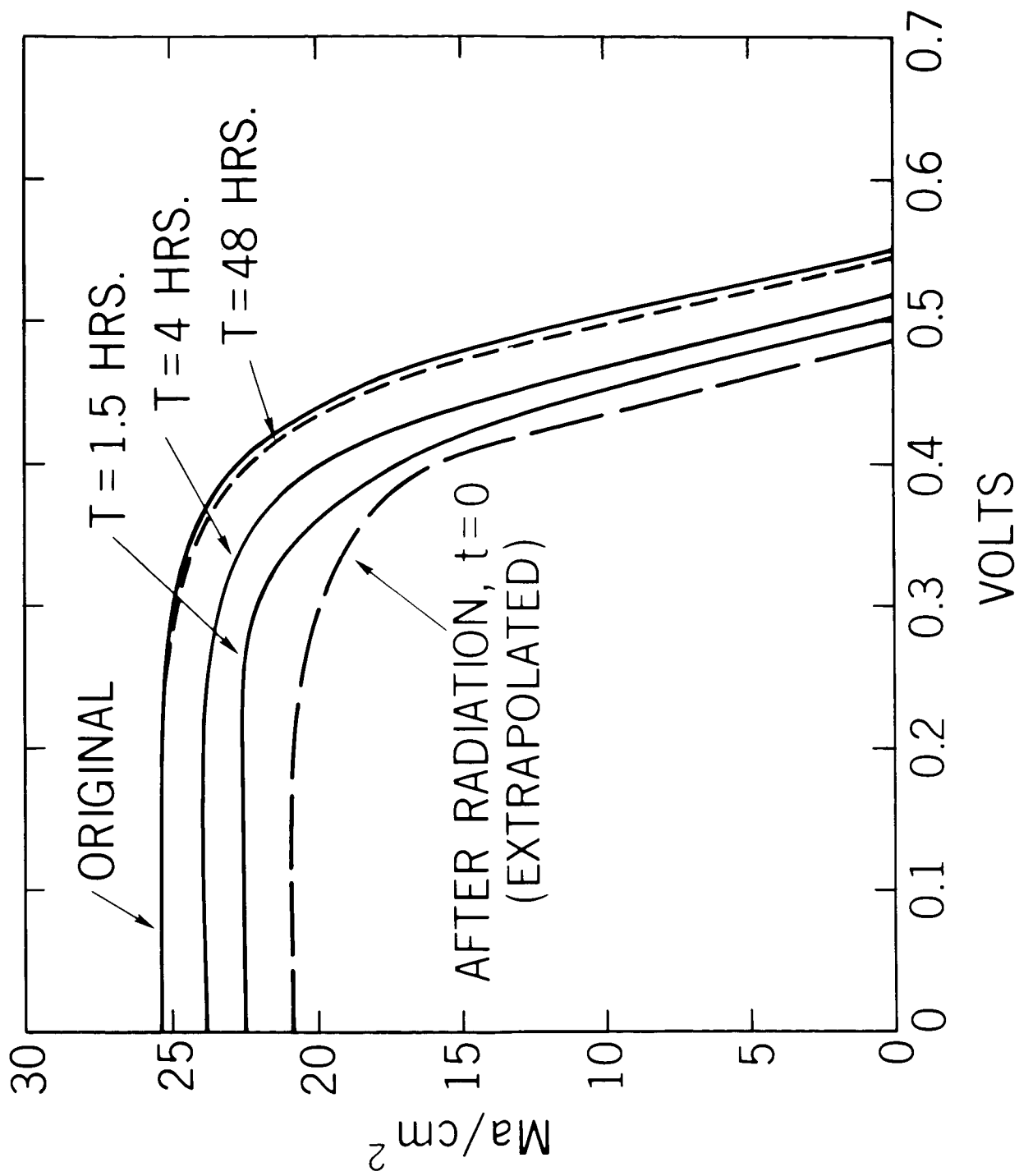
VI. RADIATION DAMAGE AND RECOVERY

The general setup for studying the photovoltaic performance of solar cells before and after radiation, and the history of recovery is similar in practice to that of the early studies of the usual n-on-p solar cells except in one aspect: In a n-on-p case, only the initial characteristics and the radiation damage are measured; the recovery, if any, will be much less than 1 percent.

In our measurement, less emphasis is placed on the assimilation of space conditions both with respect to radiation and to photo-illumination, and more emphasis is placed on a simple and repeatable measurement. In view of the great gap existing between the basic physical properties and their relation to device parameters, our philosophy has been to look at only the gross device characteristics. Specific basic studies are made only when some significant effects are recognized. In this way, information for practical purposes is provided. At the same time, in recognition of the great complexity of the electronic device — the solar cell in the present case — as a physical system, unless the effect of degradation of the recovery is large, it is almost meaningless to attempt to describe an effect as significant, which might be extraneous.

The equipment and experimental routine for the above investigation are quite simple and have been described elsewhere (Reference 19). The task in all the measurements is to compare solar cells made by different processes or irradiated under different conditions.

The primary investigation of the radiation damage in the Wysocki cell uses 1 Mev electron irradiation at room temperature. Unless otherwise specified, this will always be the condition of radiation. When the current-voltage (i-v) curve is measured for the cell before and after irradiation, typical data like those of Figure VI-1 are obtained. The i-v curves in the figure represent isothermal recovery from an initial damage which is shown as dotted lines:

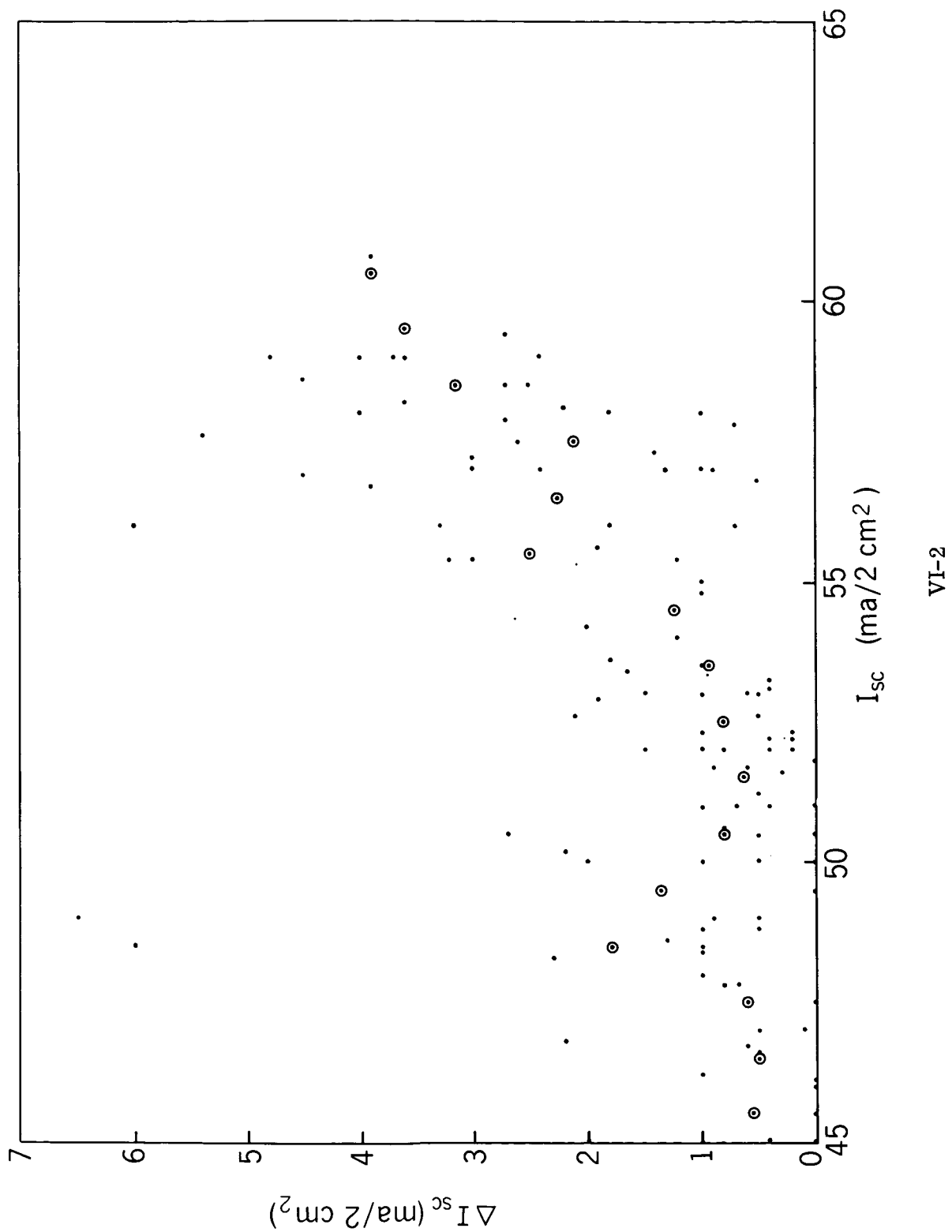


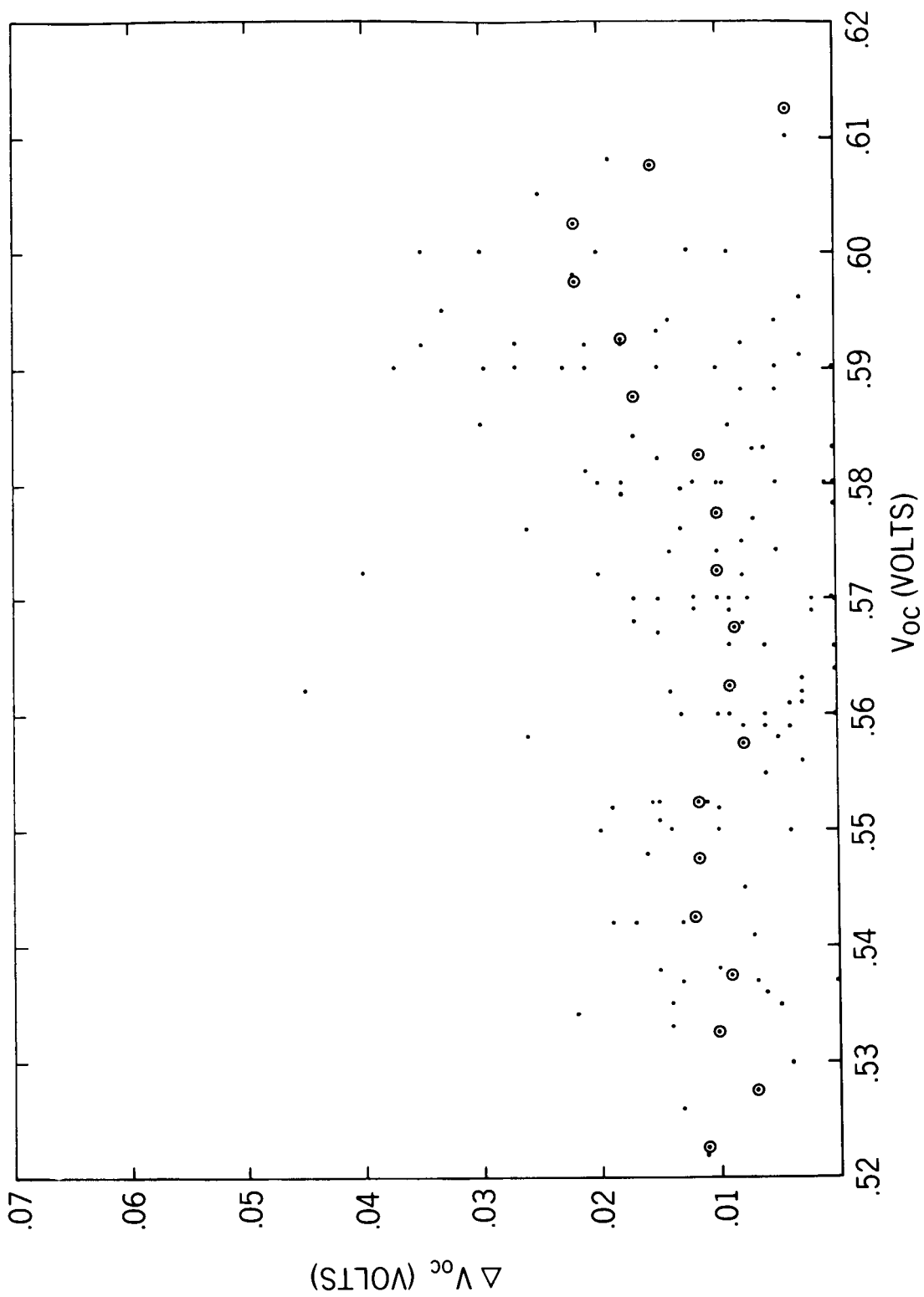
For the practical reason that the location of the radiation source and the site where the measurements were made is not contiguous, the points in the dotted line, were obtained by extrapolating the isothermal data back to zero time. From this, we estimated the initial damage as of the order of 20-percent after a 1 Mev, 10^{14} e/cm² irradiation. This damage is comparable to that of the ordinary p-on-n (no Li) solar cell and is considerably heavier than that of the ordinary n-on-p solar cell after a similar irradiation.

The characteristic time in the recovery process is on the order of hours. Since the rate of electron irradiation is 1×10^{16} e/cm² hr, the radiation time for an accumulated flux of 1×10^{14} e/cm², which is less than 1 minute, is negligibly in comparison with the recovery time. When we study the case of large flux damage, more complications arise, and additional considerations will be made.

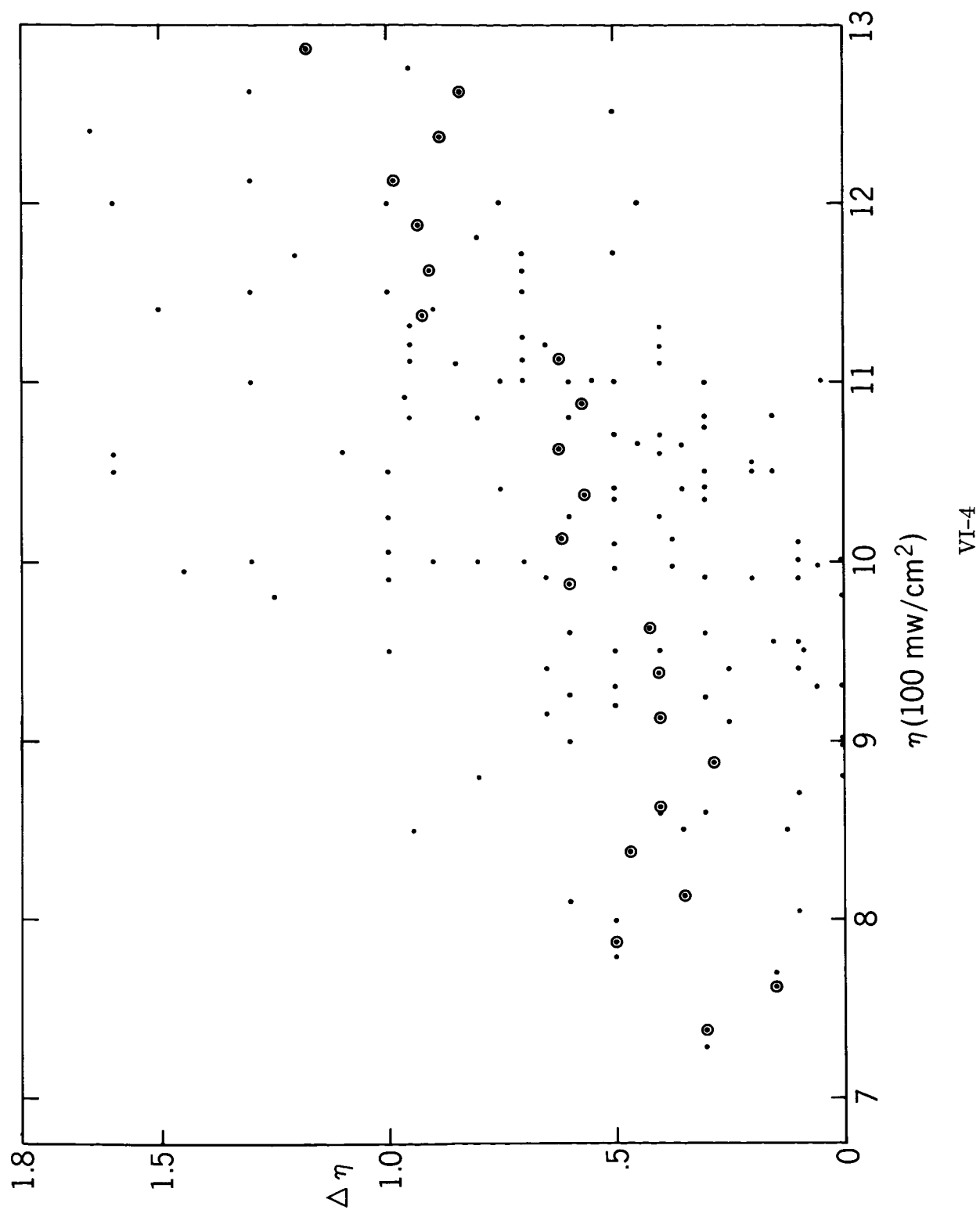
Therefore, in the first study we will confine ourselves to low flux levels of electron radiation. In the development of the analysis, more specific aspects such as the dependence on the temperature, the flux level, the flux rate, and the defect state will be discussed separately.

Solar cells made by the best methods presently available, with some variation of the temperature and the time of diffusion were received from the contractors (Reference 15) at different times and irradiated with 3×10^{14} e/cm² of 1 Mev electrons. Recoveries were measured periodically up to several weeks in some cases. From the data, we obtained histograms (Figures VI-2, VI-3, and VI-4) of the relation between the initial performance characteristics and the degradation due to the radiation of 115 solar cells. This statistical result is preliminary. At a later stage, when processes can be better controlled, a more specific analysis will be made. The histogram shows the value after degradation and recovery as a function of the original value of the short circuit current (i), open circuit voltage (v), and efficiency (η).





VI-3



There is a slight correlation of a general increase of unrecoverable damage as the photovoltaic parameters of i , v and n are increased. But there is also evidence of a considerable scattering of data which leaves a question unanswered: Whether the correlation is an intrinsic property of the Wysocki solar cell, or whether it merely represents a transitory technological problem. This question may soon be answered when large numbers of cells are produced under better controlled conditions. At present, the physical evidence does not provide a definitive correlation, at least in the low flux level case considered here.

VII. DEFECT CONCENTRATION DEPENDENCE OF DEFECT INTRODUCTION RATE

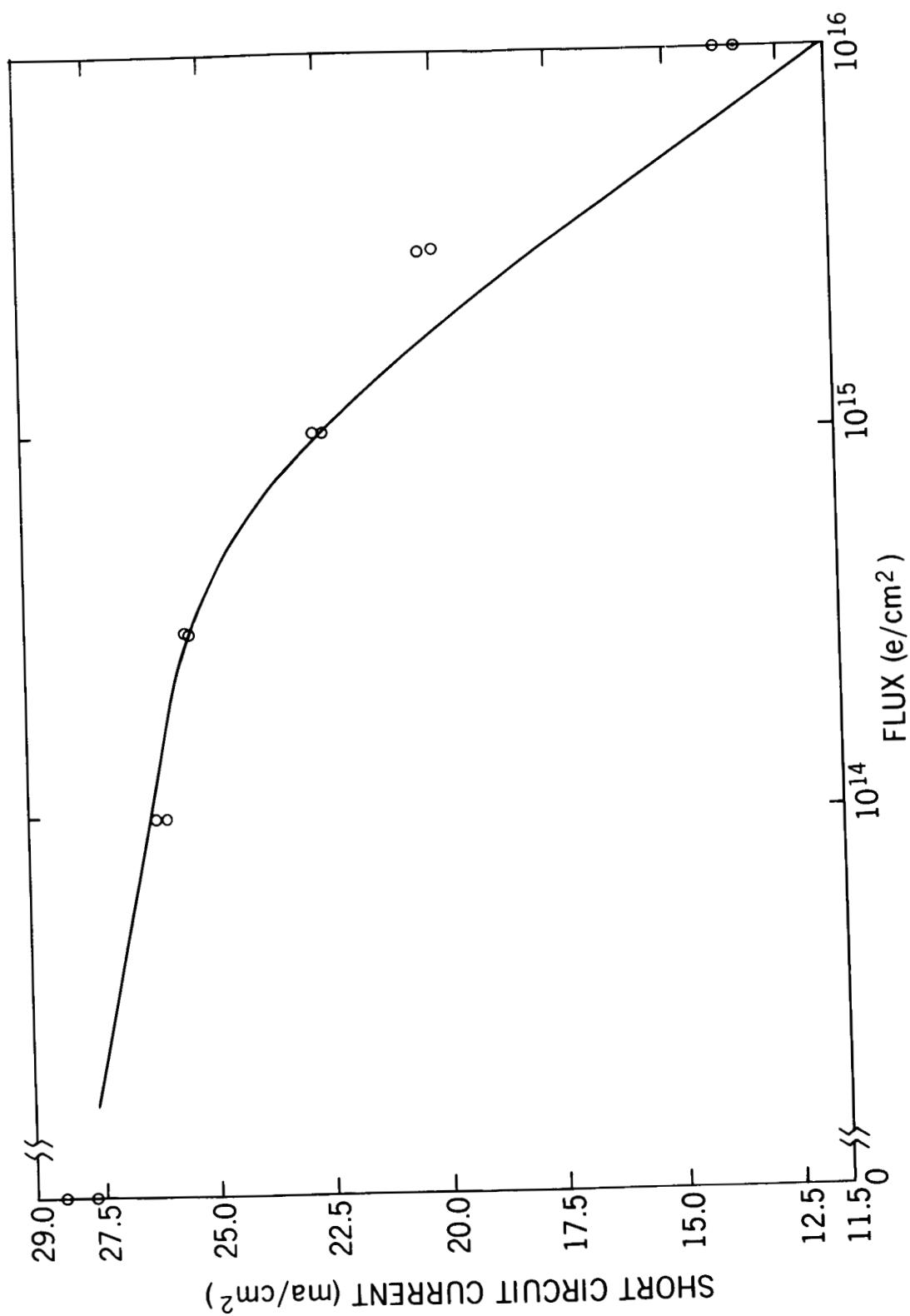
The study of the defect introduction rate in the Wysocki cell is complicated by the simultaneous occurrence of annealing. Two simplified experiments can be made:

1. The rate of defect introduction is much greater than the rate of defect annealing, and
2. The reverse of 1.

The van de Graaff irradiation, at a rate of 10^{15} to 10^{16} e/cm² -hr will approximate the first experiment when a total flux of less than 1×10^{15} e/cm² is used. Beyond this total flux level, the introduction and the annealing rate become comparable. On the other hand, in ordinary space radiation conditions the situation is closer to that of the second experiment. While the actual space radiation experiment is to be carried out in the near future, another experiment which will be closer to the space radiation one is also in preparation, which will provide a prolonged period of continuous radiation by means of a γ -source or isotope source. This investigation is in progress.

Figure VII-1 shows the radiation damage of 10 cells with initial spread also indicated. Sometimes the practice in the literature has been to use a single cell and irradiate it to successive high flux levels. Because there is evidence that the damage rate depends on the flux level as well as on the damaged state of the cell, fresh cells are used in each flux level experiment. The data are analyzed with the usual Equation of the degradation of the lifetime,

$$\frac{1}{\tau} = \frac{1}{\tau_0} + K\Phi, \quad \text{VII-1}$$



VII-1

where τ_0 and τ are the lifetime before the irradiation and after the irradiation and recovery, respectively, Φ is the flux in e/cm^2 , and K is an adjustable parameter. In this equation, a shift of Fermi level due to the carrier removal is neglected. Since the diffusion coefficient D and diffusion length L are related by

$$D_{\tau} = L^2, \quad \text{VII-2}$$

and L is proportional to the short circuit current i , we have

$$\frac{1}{i^2} = \frac{1}{i_0^2} + K' \Phi. \quad \text{VII-3}$$

By fitting data at $\Phi = 1 \times 10^{15}$,

$$K' = 1.51 \times 10^{-13} \text{ (cm}^4/\text{amp}^2\text{-e)}. \quad \text{VII-4}$$

Equation VII-3 is computed with this K' value and is shown as a continuous curve in Figure VII-1. The curve fitting is excellent in the low flux region. The deviation at $3 \times 10^{15} e/cm^2$ can be explained by the spontaneous recovery taking place during the irradiation, that is, the initial damage after the cessation of the radiation is lower than that which would be caused by a pulse of $1 \times 10^{15} e/cm^2$.

The same observation applies to the data of $1 \times 10^{16} e/cm^2$ radiation. In this case, an extrapolation from the data of 3×10^{15} would indicate a stronger deviation from the value predicted by Equation VII-3. The weaker deviation observed can be explained by an onset of a steeper damage introduction rate near this high flux.

To improve the analysis, a narrow band penetrative light source was used and the residual damage was analyzed. The result will be discussed in the next section.

VIII. DEFECT CONCENTRATION DEPENDENCE OF ANNEALING RATE

In the study of the annealing of radiation-induced defects in semiconductors some evidence was obtained on the dependence of the annealing rate on the concentration of impurities (Reference 20).

When the irradiation is carried out at room temperature, most of the defects formed in Si are stable up to several hundred degrees centigrade*. Therefore, isothermal annealing, on which the annealing rate is measured, has to be carried out at high temperatures. This introduces some complexities in the interpretation of the data since the activation energy of the annealing itself could depend on the temperature (Reference 21). This difficulty is minimized in the case of annealing of the defects in the Wysocki cell because in this case both the radiation and annealing occur at room temperature. Therefore, the entire experiment can be carried out at a single temperature.

Solar cells are irradiated with different total fluxes of 1 Mev electrons at a rate of 3×10^{12} e/cm² sec. The specimen is placed on a water-cooled aluminum plate, and the temperature rise during the irradiation is less than 5° C. The isothermal change of the short circuit current of the solar cell at room temperature is measured. The data are shown in Figure VIII-1.

In the initial stage of recovery, an exponential time dependence adequately describes the experiment result. We have, for the fraction of residual damage f ,

$$i_o - i_a \approx \exp(-t/\tau), \quad \text{VIII-1}$$

* R. R. Hasiguti and S. Ishino, loc. cit. Reference 6; p. 259.

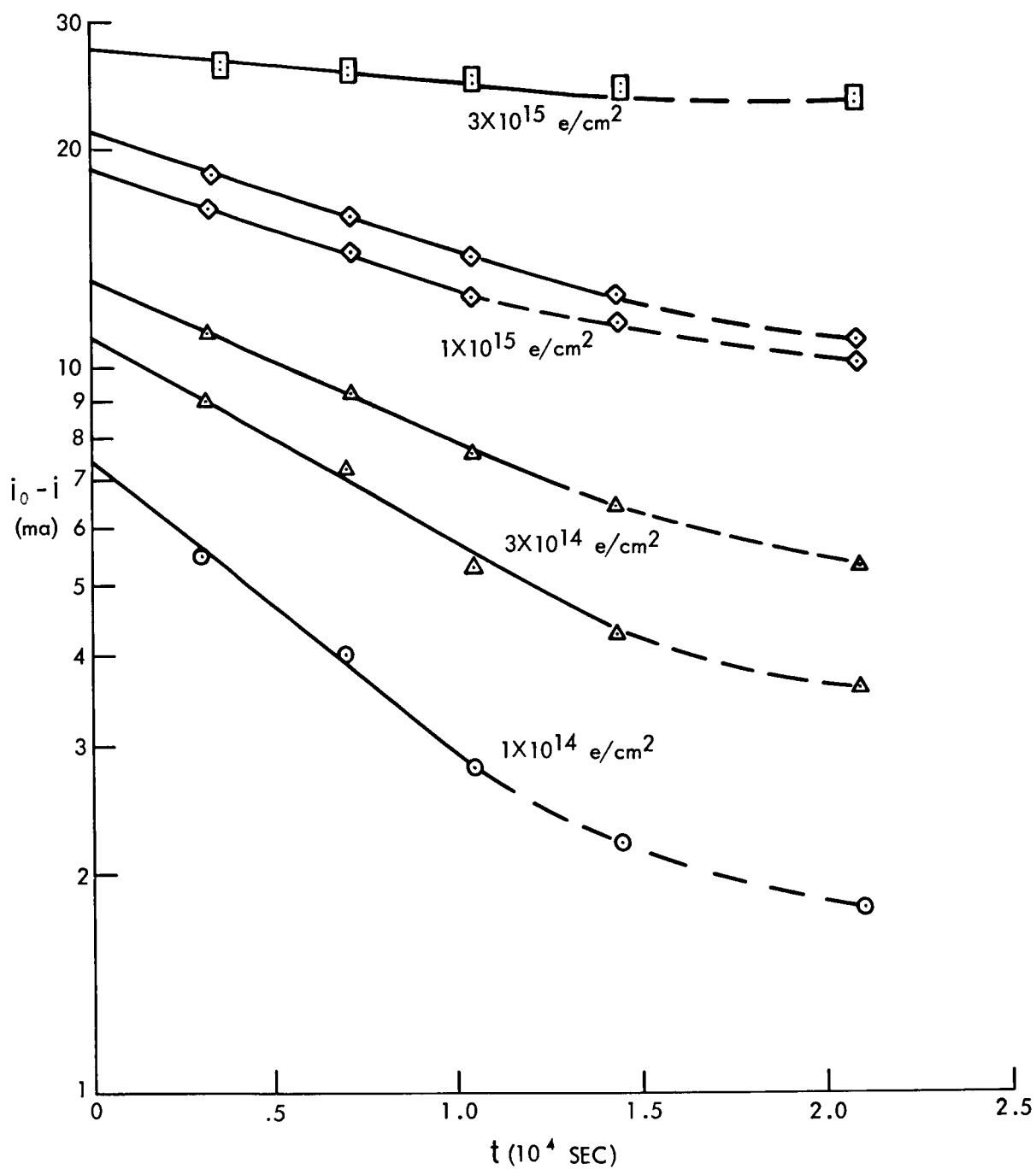


Figure VIII-1

Where i is the short circuit current, the subscripts o and a stand for initial and after annealing, respectively, and t is the time variable. The results are given in the following table. The recovery time is evaluated from the linear portion of the curves.

Table VIII-1

Specimen	i_o ma/2 cm ²	Total flux/cm ²	$\tau \times 10^4$ sec ma/2 cm ²
1	53.5	1×10^{14}	1.1
2	53.2	3×10^{14}	1.4
3	49.4	3×10^{14}	1.9
4	49.4	1×10^{15}	2.5
5	50.8	1×10^{15}	2.8
6	50.0	3×10^{15}	10.6
7	49.2	3×10^{15}	10.6

In a more extensive analysis, it is necessary to minimize the contribution of p-region degradation. This is achieved by injecting a penetrating light of 8420 Å (with 100Å bandwidth). Furthermore, a linear proportionality between the diffusion length and the short circuit current, is assumed and the life time of the minority carriers to be proportioned to L^{-2} therefore, the fraction of the annealed defects is defined as

$$f = (i_a^{-2} - i_o^{-2}) / (i_{\Phi} - i_o^{-2}), \quad \text{VIII-2}$$

where i_{Φ} is the short circuit current after irradiation with a total flux Φ .

In the computation of f , we have subtracted a portion of the photovoltaic current i_c from i_o . The current i_c represents the difference between i_o and i_a measured after a very long time when no further change in i_a is observed.

This portion of i obviously is contributed by other types of defects and therefore is subtracted in our analysis.

Figure VIII-2 shows the isothermal annealing data of f for specimens with different total fluxes of irradiation. We observe a shift of the isothermal annealing curve towards the longer time. This corresponds to an increase of the recovery time as the defect concentration is increased.

The important implication of this result is that the dependence of the recovery rate on the defect concentration is opposite to the prediction of current models which can be collectively represented by (Reference 22)

$$f = \frac{1}{1 + \lambda N_{\Phi} \tau}, \quad \text{VIII-3}$$

where N_{Φ} is the concentration of defects and λ is a positive parameter independent of Φ . From the dimensional structure, therefore $(\lambda N_{\Phi})^{-1}$ is a measure of the recovery time. Since N_{Φ} is a positive increasing function of Φ , Equation VIII-2 predicts a decrease of the recovery time with respect to the concentration of defects.

Taking an arbitrary level of f , we obtain the concentration dependent recovery time τ which is given in Figure VIII-3. For values of τ at $f = 0.50$, a good approximation is obtained by

$$\tau_{0.5} = 74.5 \exp (2.86 \times 10^{-5} \Phi^{1/3}). \quad \text{VIII-4}$$

On the other hand, for values of τ at $f = 0.10$,

$$\tau_{0.1} = 5.06 \times 10^2 \exp (2.73 \times 10^{-5} \Phi^{1/3}). \quad \text{VIII-5}$$

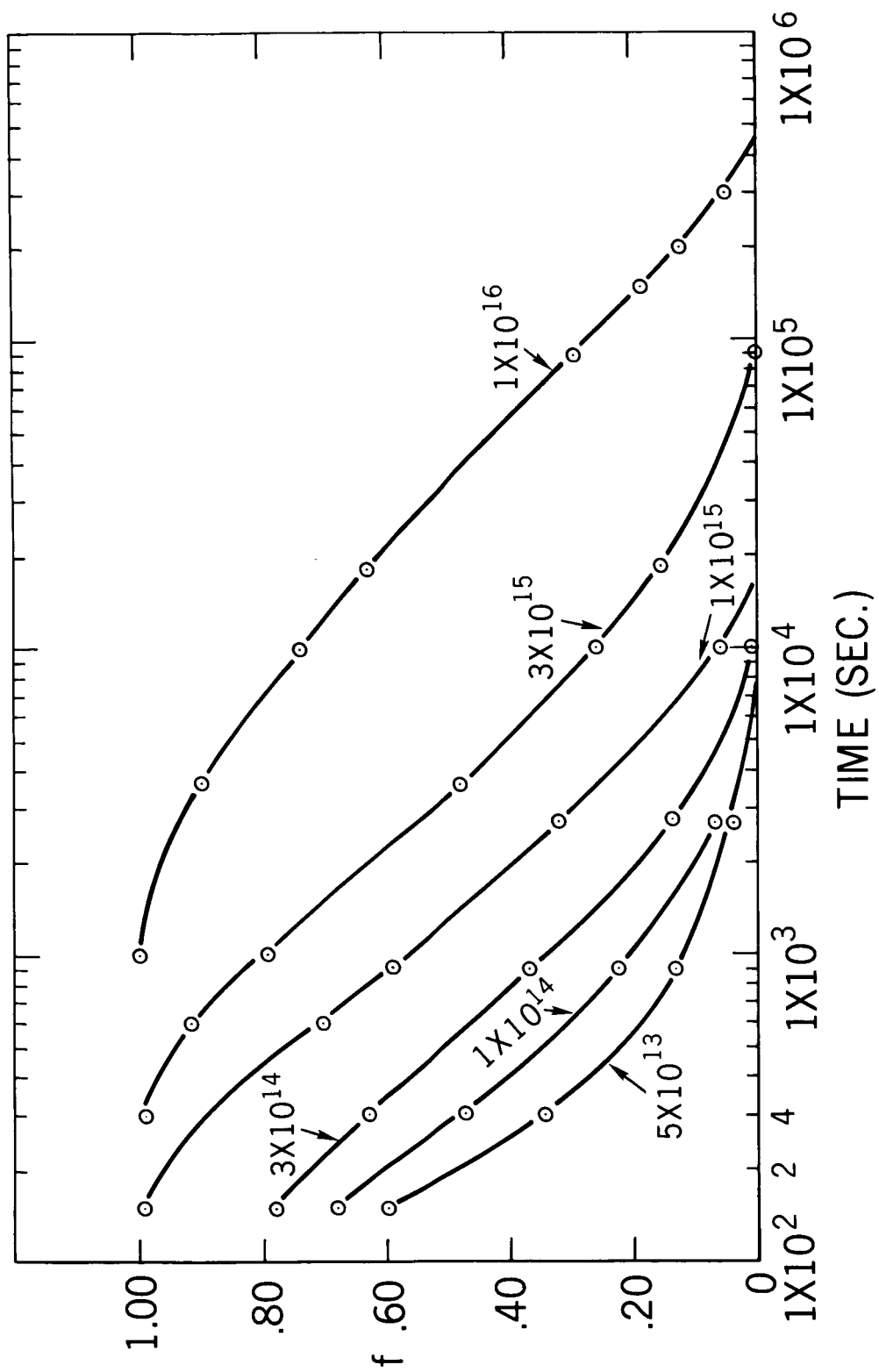
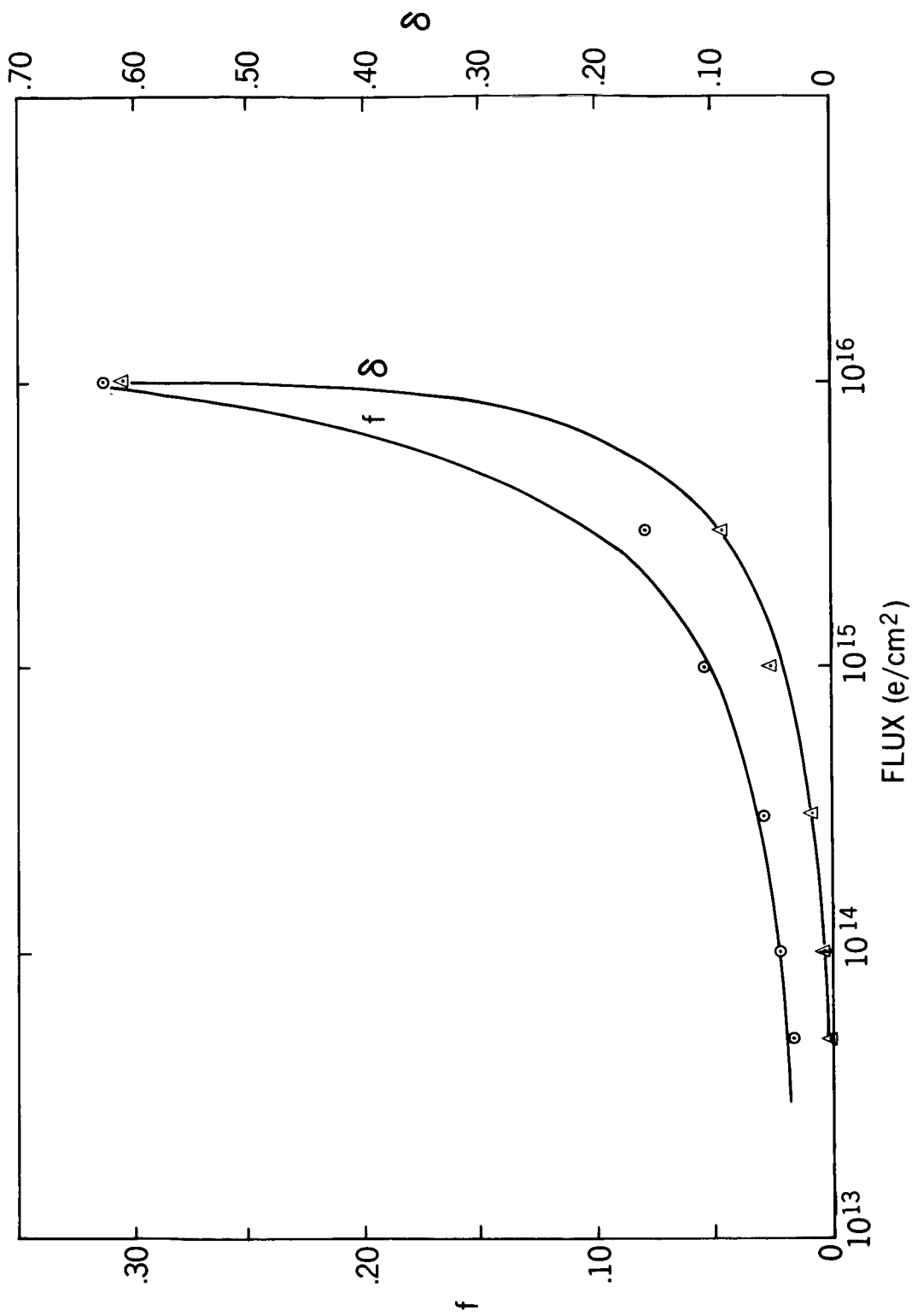
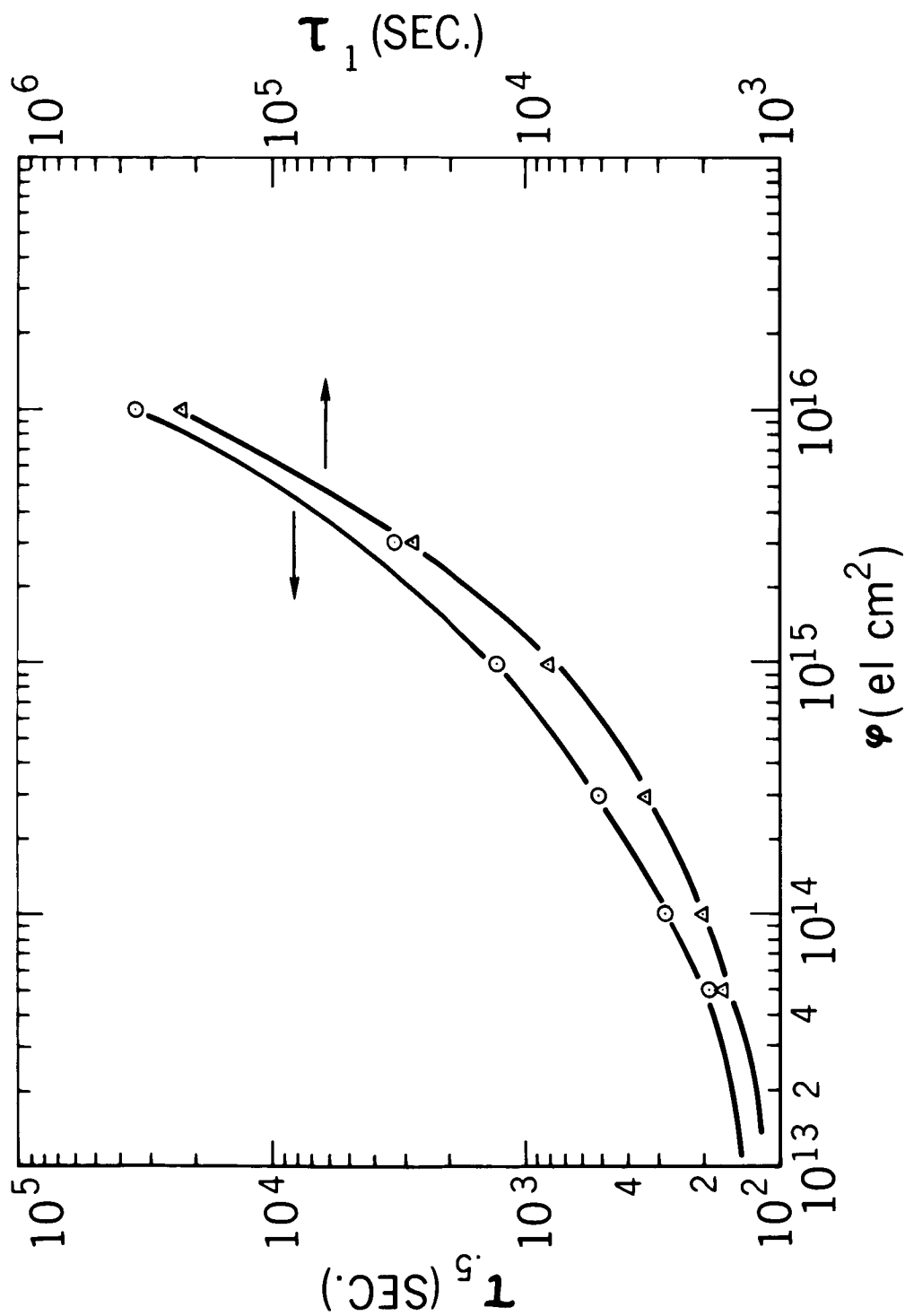


Figure VIII-2



VPI-3



VIII-4

It is interesting to note that the main difference between Equations VIII-4 and VIII-5 is the pre-exponential factor. The numerical values given in Equations VIII-4 and VIII-5 could conceivably be dependent upon the processes and the detailed technology of the solar cell fabrication. The importance is that a $\Phi^{1/3}$ dependence describes well the general behavior.

From the same data, we can analyze the dependence of the fraction of the residual damage on the concentration of the defect. Figure VIII-4 shows the result. The curve is calculated from the following equation:

$$f = 6.38 \times 10^{-3} \exp (2.12 \times 10^{-5} \Phi^{1/3}). \quad \text{VIII-6}$$

In the same figure, $\delta = i_{\Phi}^{-2} - i_0^{-2}$ is also shown. The same $\Phi^{1/3}$ dependence is also found to be a satisfactory description of the experimental data.

Within the experimental accuracy, the coefficients in the exponential can be treated as if they were about the same.

We will return to this discussion in the section entitled "Kinetics of Annealing."

IX. KINETICS OF ANNEALING

In the early investigation of the annealing, the experimental data used in making comparisons with physical models was exclusively that obtained from isothermal and isochronal annealing. (Reference 23). Our investigation shows that the flux dependence of the annealing provides a more stringent test of the physical model. By this test, we have proved that currently existing annealing models predict annealing properties in contradiction to the experimental observations. A phenomenological model is proposed* in which an interaction is assumed between defects, and consequently the activation energy of defect formation and annihilation becomes a function of the defect concentration.

Since the above work has already been reported in the literature, a repeated description, even if it could have some pedagogical value in constructing some mathematical approved--the original papers are necessarily brief because they appear as letters--will not be attempted here. At this stage, a more important problem is to look for the physical foundation of this model.

First, we will estimate the activation energy represented by Equation VIII-5:

$$\tau_{0.1} = 5.06 \times 10^2 \exp 2.73 \times 10^{-5} \bar{\phi}^{1/3} \text{ sec.} \quad \text{IX-1}$$

Since the experiment pertains to room temperature (300°K), we have

$$2.73 \times 10^{-5} \bar{\phi}^{1/3} = \frac{11610}{300} E_1, \quad \text{IX-2}$$

where E_1 is the defect concentration dependent part of the activation energy.

*P. H. Fang, Phys. Letters. 24 A, (251) 1967.

In this sense,

$$\tau_0 = 5.06 \times 10^2 \exp (-E_0/k T), \quad \text{IX-3}$$

where E_0 is independent of concentration. From the discussion in the section entitled "Li-Solar Cell Technology," we will tentatively assign

$$E_0 = 0.66 \text{ ev.} \quad \text{IX-4}$$

Therefore, $\tau_0 = 1.01 \times 10^{-9} \text{ sec.}$ On the other hand,

$$E_1 = 7.06 \times 10^{-7} \Phi^{1/3} \quad \text{IX-5}$$

The defect production rate in n-type Si is on the order of 0.1 to 0.01. In this case,

$$E_1 = (1.52 \text{ to } 3.29) \times 10^{-7} n^{1/3}, \quad \text{IX-6}$$

where n is the defect density. Therefore, for $n = 10^{12}$, $E_1 = 1.52 \text{ to } 3.29 \text{ mv}$; for $n = 10^{15}$, $E = 15.2 \text{ to } 32.9 \text{ mv}$. This is quite small compared with the E_0 value of 660 mv; therefore, the E -value will dominate in the measurement of the activation energy.

In the search for a possible mechanism, we note that in the case of Hall conductivity in germanium (Ge) an impurity concentration dependent activation energy is given by (Reference 24, 25),

$$E = E_0 - (2.35 \text{ to } 4.3) \times 10^{-8} n^{1/3} \text{ ev.} \quad \text{IX-7}$$

The numbers involved are not very different. The positive sign in the case of Equation IX-6, versus the negative sign in the case of Equation IX-7, for the

term $n^{1/3}$ could be attributed to the increase or the relief of the strains in the lattice respectively.

Finally, according to the previous work*, we have the following kinetic equation:

$$\frac{dn}{dt} = \frac{n}{502} (-E_1/kT). \quad \text{IX-8}$$

After integration and taking $n = n_0$ at $t = 0$,

$$\frac{1}{E_1} (\alpha n_0^{1/3}) - \frac{1}{E} (\alpha n^{1/3}) = t/1506. \quad \text{IX-9}$$

This will be the isothermal equation. The value of α is given by Equation IX-6 and is dependent on the temperature.

*P. H. Fang, Phys. Letters 24 A, (251) 1967.

X. DYNAMICS OF ANNEALING

The problem to be discussed is the situation one would encounter in space applications. In view of the scarcity and the tentativeness of the available data, the point to be stressed is the qualitative characteristics rather than the actual numerical value of the computations.

Based on the equation of the kinetics of annealing, if there is a simultaneous recovery in the presence of a radiation source with a flux of v electrons per second, the complete equation of dynamical balance becomes

$$\frac{dn}{dt} = \frac{n \exp(-\alpha n^{1/3})}{\tau_0} + v, \quad \text{X-1}$$

where the numerical values of α and τ_0 are given by Equations IX-1 and IX-6, respectively. In the case of $v = 0$, Equation X-1 can be solved in closed form as given by Equation IX-9. In the case of $v \neq 0$, numerical methods have to be applied. However, an analysis can be made with the following asymptotic method.

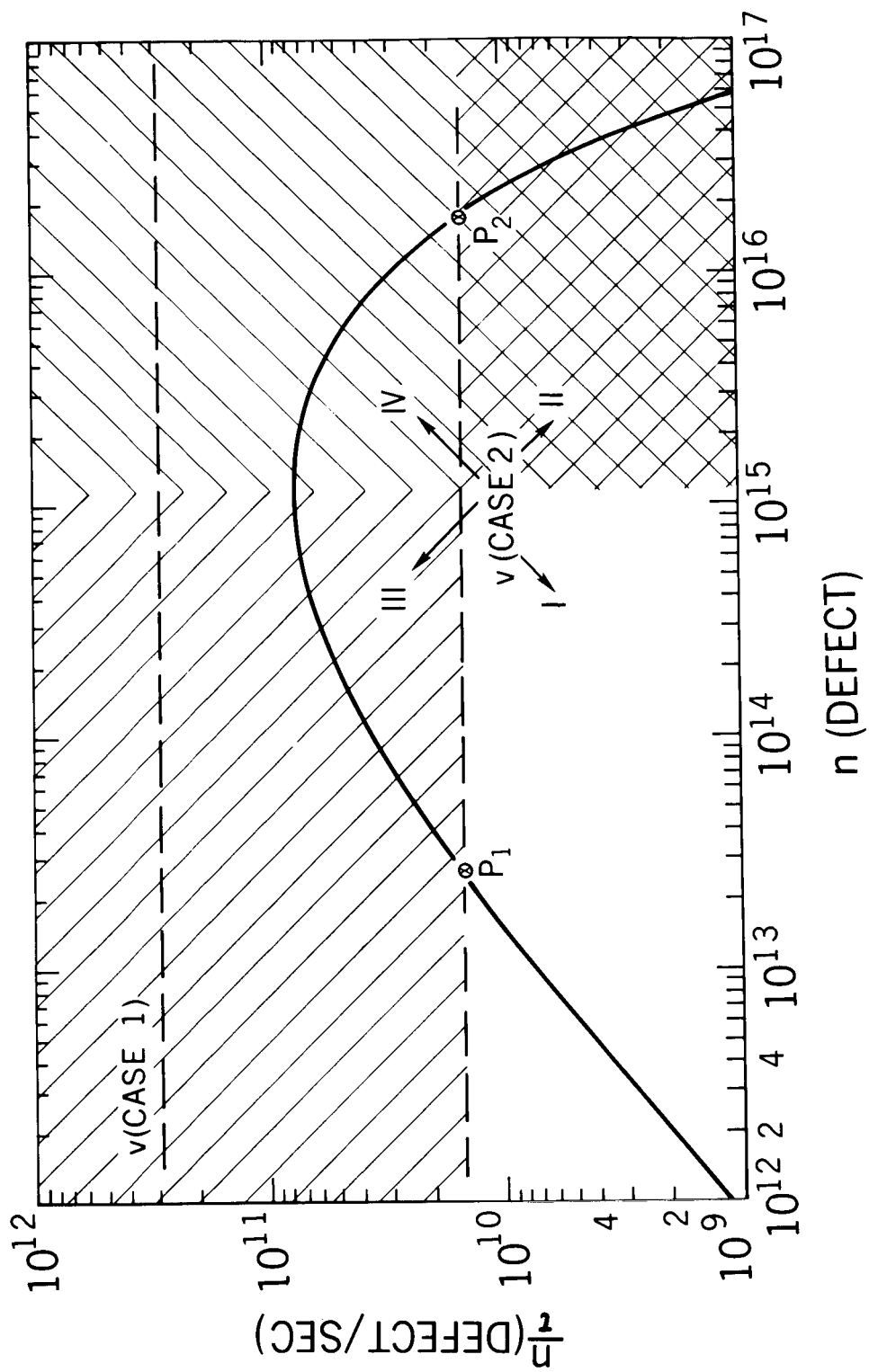
Expanding the first term of the right side of Equation X-1 about a point n_0 - corresponding to the initial value of the defect density, Equation X-1 becomes approximately $(\lambda - 1/\tau_0)$, and

$$\begin{aligned} \frac{d\mu}{dt} &= - [(v - \lambda n_0 \exp(-kn_0^{1/3})) - \lambda(1 - \frac{k}{3} n_0^{1/3}) \exp(-kn_0^{1/3})] \mu \\ &= A - B\mu, \end{aligned} \quad \text{X-2}$$

where $\mu = n - n_0$. We therefore obtain

$$\mu = \frac{A}{B} (1 - e^{-Bt}).$$

When $B > 0$, the time development is a slow process, but when $B < 0$ the time development is rapid.



X-1

We have the following table, where the regions are shown in Figure X-1 for case 2.

Region	A	B	Characteristic
I	+	+	Slow increase in damage
II	+	-	Fast increase in damage
III	-	+	Slow annealing
IV	-	-	Fast annealing

The solid curve in Figure X-1 represents n/τ for various values of n where v is larger than a maximum of n/τ . The four regions are represented by different shaded areas. It is interesting to see that in region II, which represents a heavy initial damage, as time progresses, even at moderate irradiation rates the damage will increase further.

In case 1, where $v = \max (n/\tau)$, regions III and IV vanished. Therefore, no apparent annealing would be observed. On the other hand, if v is very small, regions III and IV will be expanded, that is, the annealing region will be dominant.

The treatment here has been confined to the case where v does not change as a function of the time. In general, v is not a constant and the initial defect density has to be treated as a movable initial condition. The computation will be very complicated, but the principle involved is well illustrated by the present discussion.

XI. SOME EXPERIMENTAL RESULTS PERTAINING TO SPACE APPLICATIONS

Some essential ingredients for simulating space conditions, from the point of view of radiation damage in solar cells, are:

1. A spectrum of energetic rays and particles, mostly electrons and protons, with a moderate rate of about 10^{11} to 10^{13} particles/cm²-day.
2. An ultrahigh vacuum.
3. A system temperature from -100° to 50° C.

In performing the experiments, for ordinary solar cells which have no significant annealing effects in the space environment, the radiation damage of solar cells depends only on the total radiation dose, but not on the dose rate. Therefore, the true function of the radiation $v(t)$ is not important. As discussed in the section entitled "Dynamics of Annealing", in the case of the Li solar cell $v(t)$ is of fundamental importance. The status of the present knowledge of some basic parameters required to predict the damage behavior for a given $v(t)$ is such that even a semiquantitative estimation cannot be made. This implies the importance of obtaining information regarding the energy spectrum and dose rate of the radiation source.

A high vacuum condition could be important and beneficial especially in the case of low energy irradiation. In a special experiment carried out in an ambient atmosphere, we observed that a prolonged radiation for several weeks on solar cells definitely leads to a discoloration of the silicon oxide antireflecting surface. This is caused by the surface bombardment by gas ions originating in the atmosphere.

The temperature effect of the radiation damage and annealing in the Li solar cell has not been studied sufficiently. Some initial observations imply that

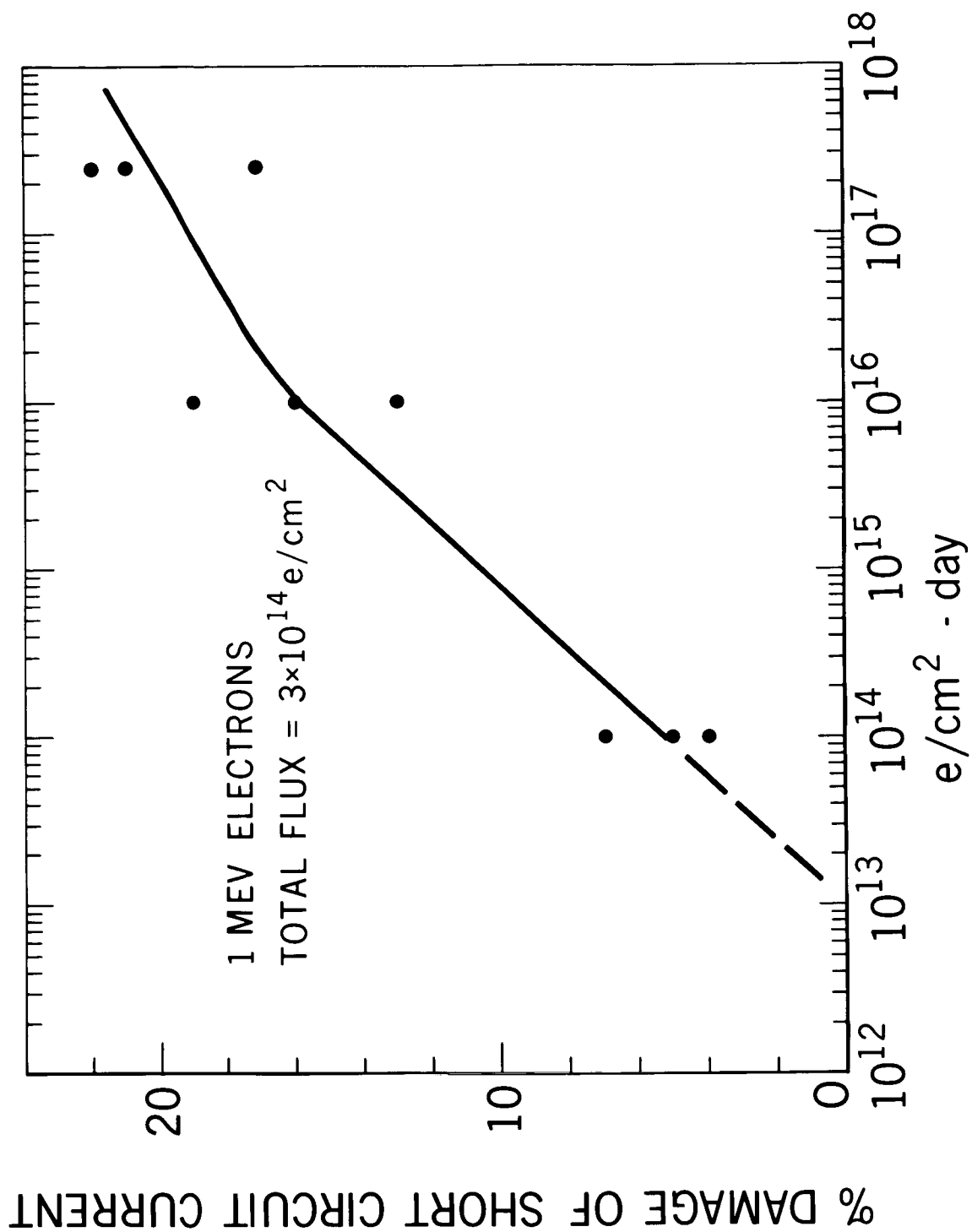
the high damage introduction rate occurs at a temperature of -50°C or below. At temperatures higher than 70°C , a simultaneous annealing is so effective that no appreciable damage is observed.

Some special experiments already carried out or now underway will now be discussed:

1. Dose rate dependent experiment. In this experiment, van de Graaff irradiations of 1 Mev electrons of three flux rates are used, 1×10^{14} , 1×10^{16} , and 2.4×10^{17} e/cm²-day. The total accumulated flux is 3×10^{14} e/cm². Degradation of the short circuit current of three groups of solar cells, each group consisting of three, is measured 1 hour after the cessation of the radiation (Figure XI-1).

First, we observe an increase of the residual damage as the flux rate is increased. An extrapolation indicates a vanishing residual damage if the flux rate is less than 1×10^{13} e/cm²-day. This is generally satisfied in the actual space environment.

Second, the data presented were for the residual damage measured 1 hour after the cessation of the radiation. A further annealing to complete recovery is obtained if two additional hours are allowed to lapse in the 1×10^{14} e/cm²-day radiation case, while it requires 5 days for the 2.4×10^{17} e/cm²-day case. Therefore, a slow radiation rate results in less damage; also, slow radiation requires less time for recovery even when "time" is a sum of radiation time and annealing time.



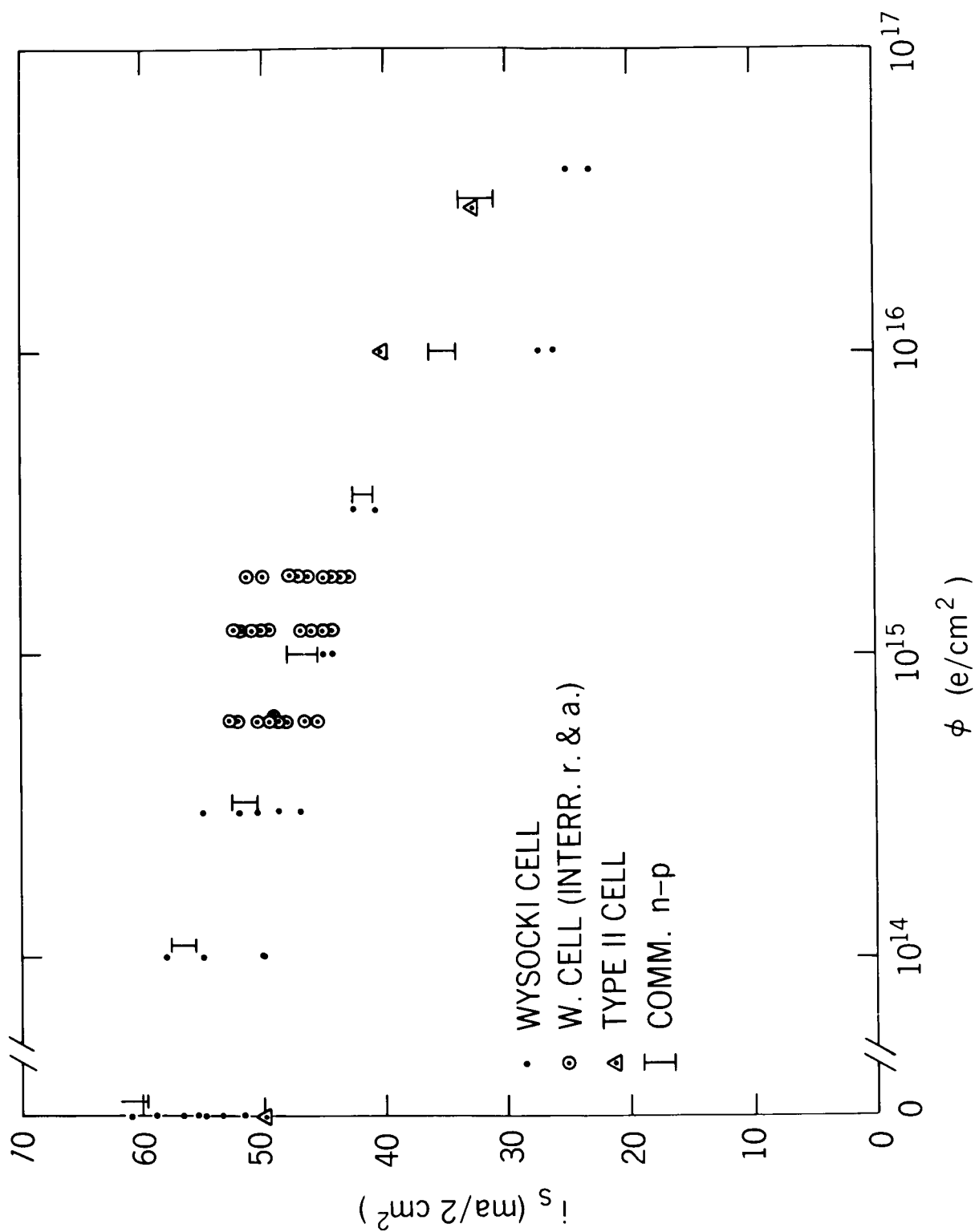
XI-1

The data of Figure XI-1 are obtained from solar cells fabricated 3 months ago. The experiment will be repeated with more recent solar cells in hopes of obtaining better statistics from an improved type of solar cell.

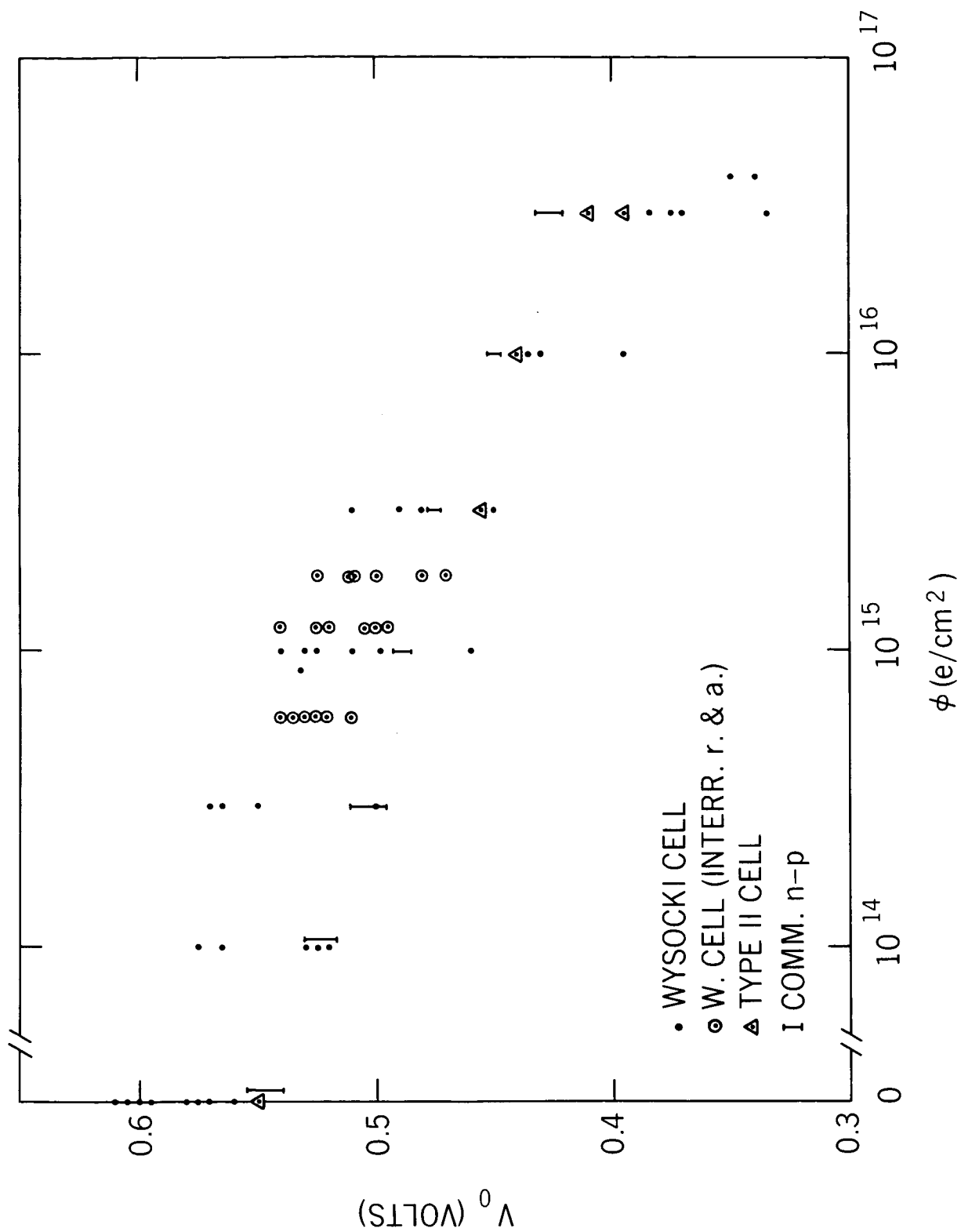
2. Repeated and interrupted radiation. In this experiment, a group of solar cells is irradiated with a flux rate of $1 \times 10^{16} \text{ e/cm}^2\text{-hr}$ to the desired level of accumulated flux (solid points in Figures XI-2, XI-3, and XI-4). The second group is irradiated at the same rate to $3 \times 10^{14} \text{ e/cm}^2$ (circular points in Figures XI-2, XI-3, and XI-4) and is repeated each time after about 10 days. The results show that the degradation in the case of repeated radiation and interruption, for an equal accumulated flux, is smaller than in the case of a continuous radiation. In the same figure, a few triangular data points are given. This data represent the Type II Li cell which was described in the section entitled "Different Types of Li Solar Cells". At high accumulated flux levels, the Type II cell definitely shows a lower level of degradation. But in the case where the defect introduction rate is not excessive such a type of solar cells, which represents a somewhat complex construction, is probably not necessary.

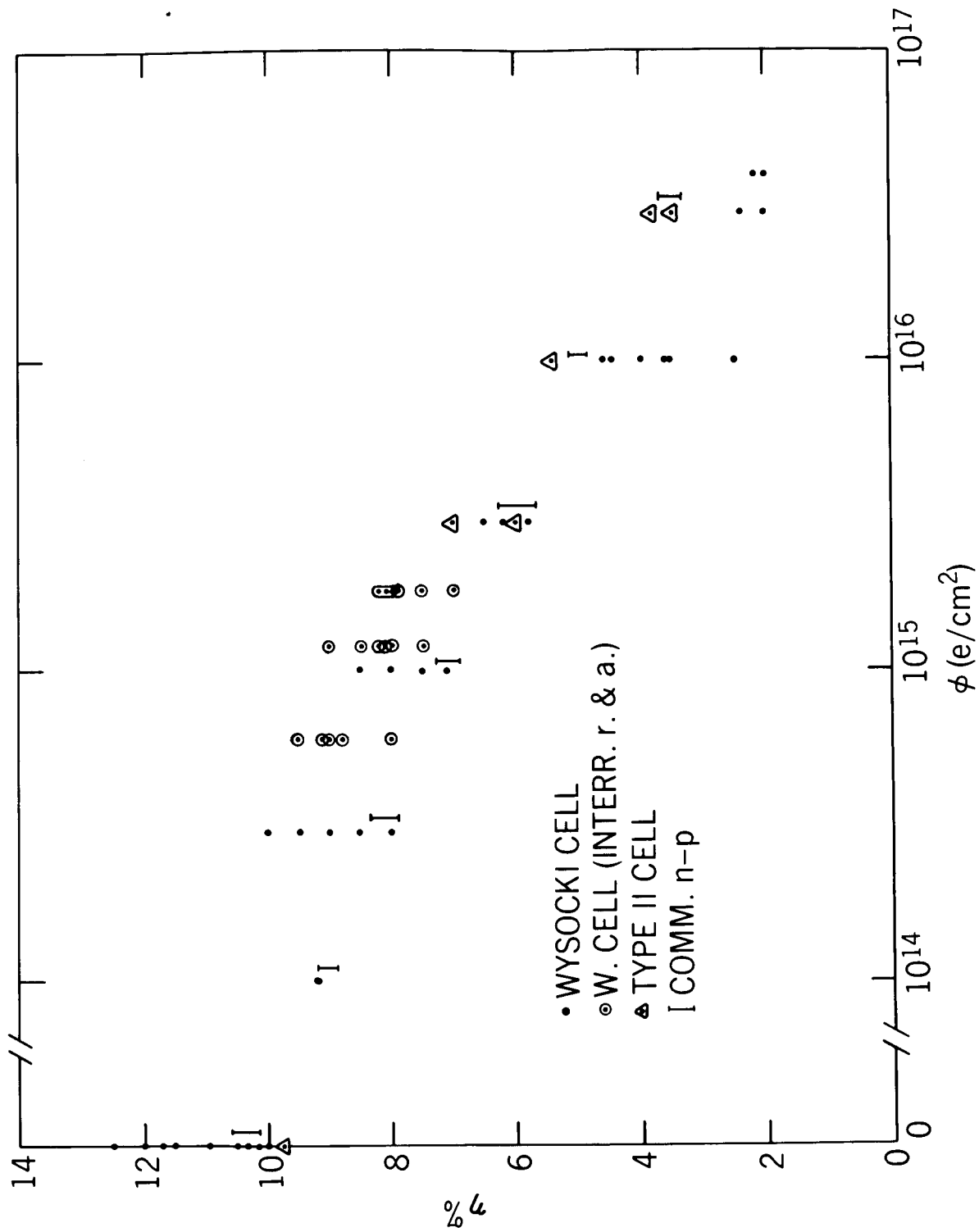
Although Figures XI-2, XI-3, and XI-4 give the statistical distribution of the experimental data, a simple arithmetic average, which gives a somewhat clearer picture, is given in Figures XI-5, XI-6, and XI-7.

3. Some other experiments in prospect are: (a) Use a $\text{Sv}^{90} - \text{Y}^{90}$ isotope source for β -rays and irradiate at a level of $1 \times 10^{12} \text{ e/cm}^2\text{-day}$ in vacuum conditions at different temperatures; (b) determine the stability of the Li cell at different temperatures with different damaged states; and (c) determine the effect of photon injection on the rate of defect introduction and annealing.

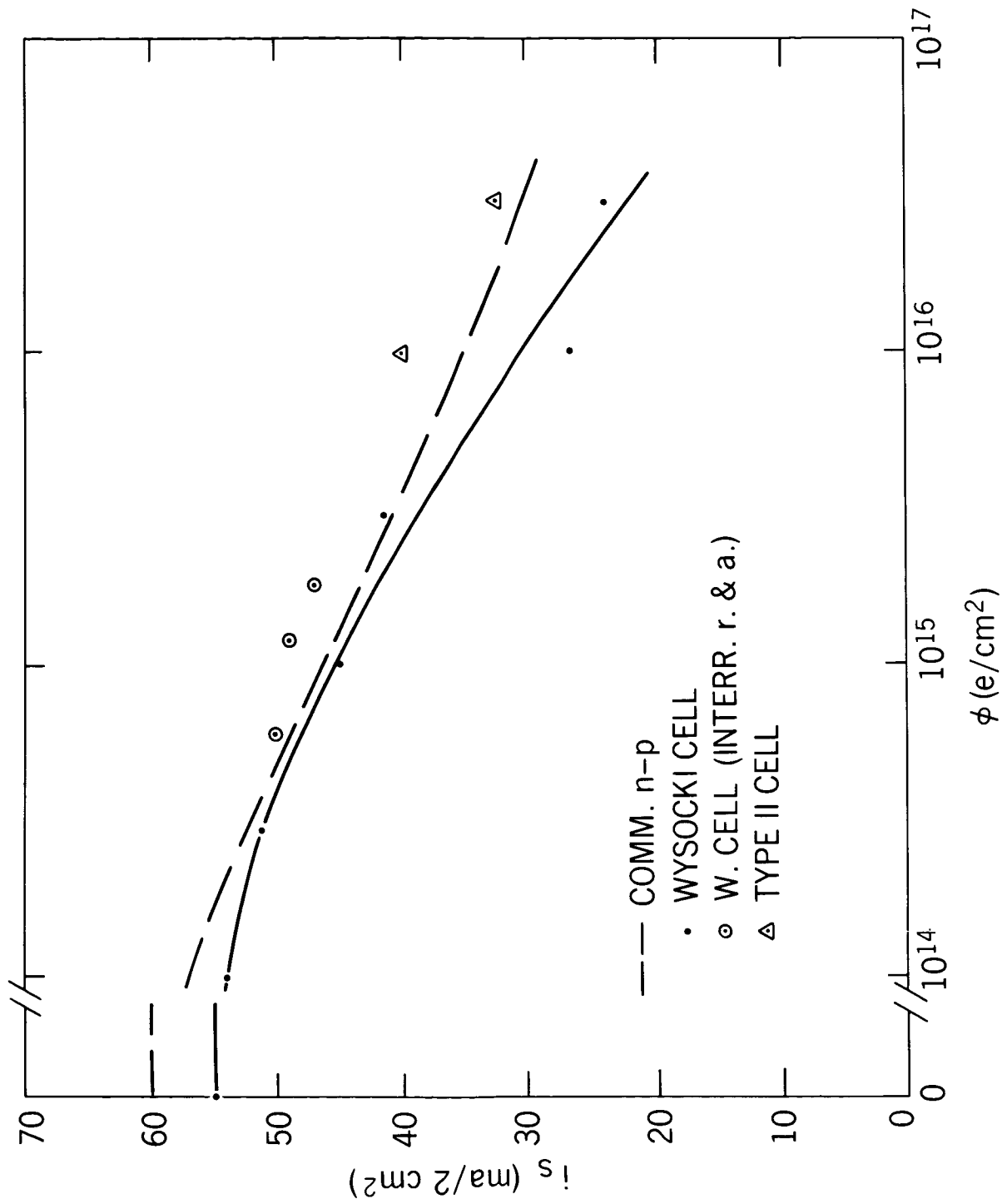


XI-2

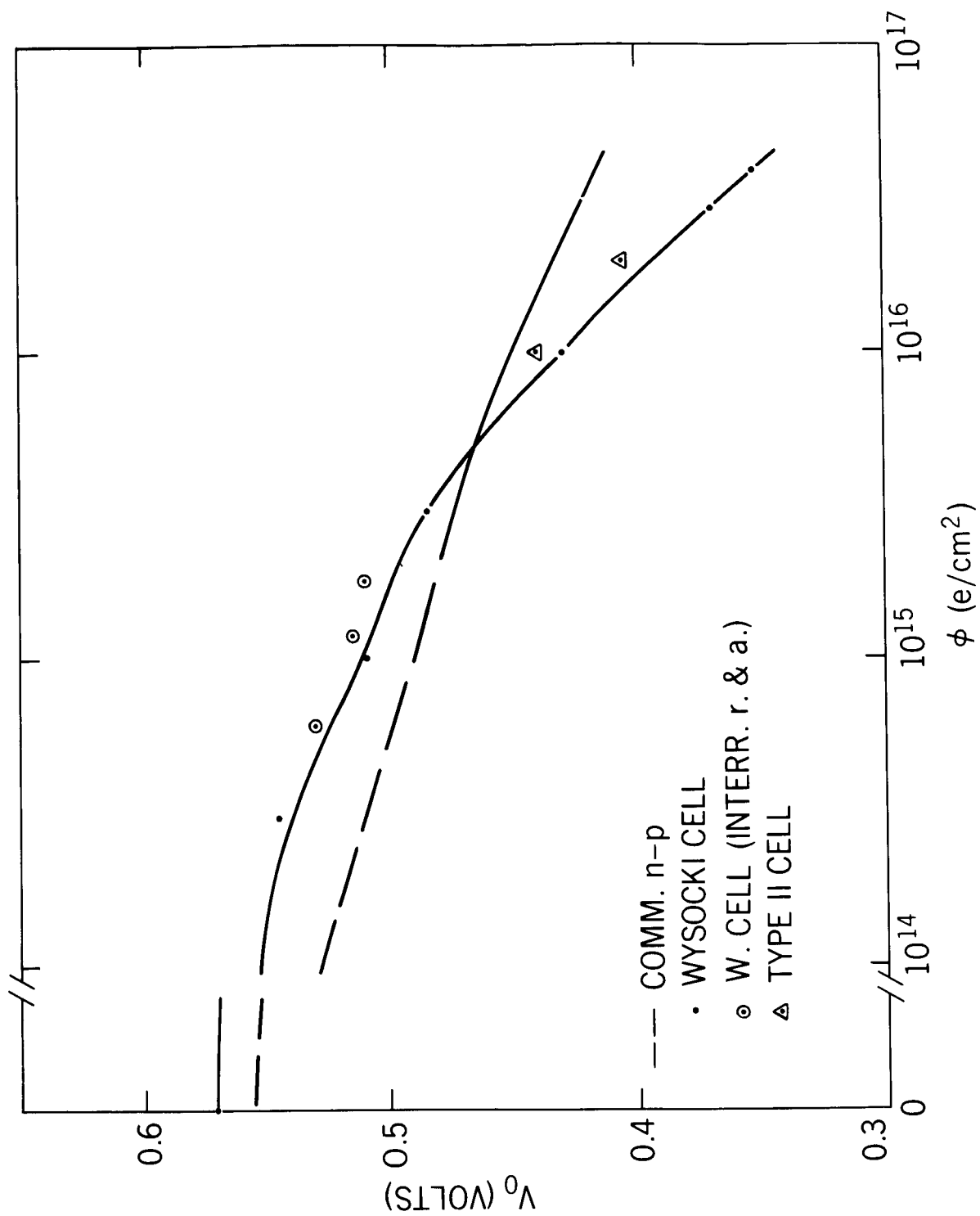




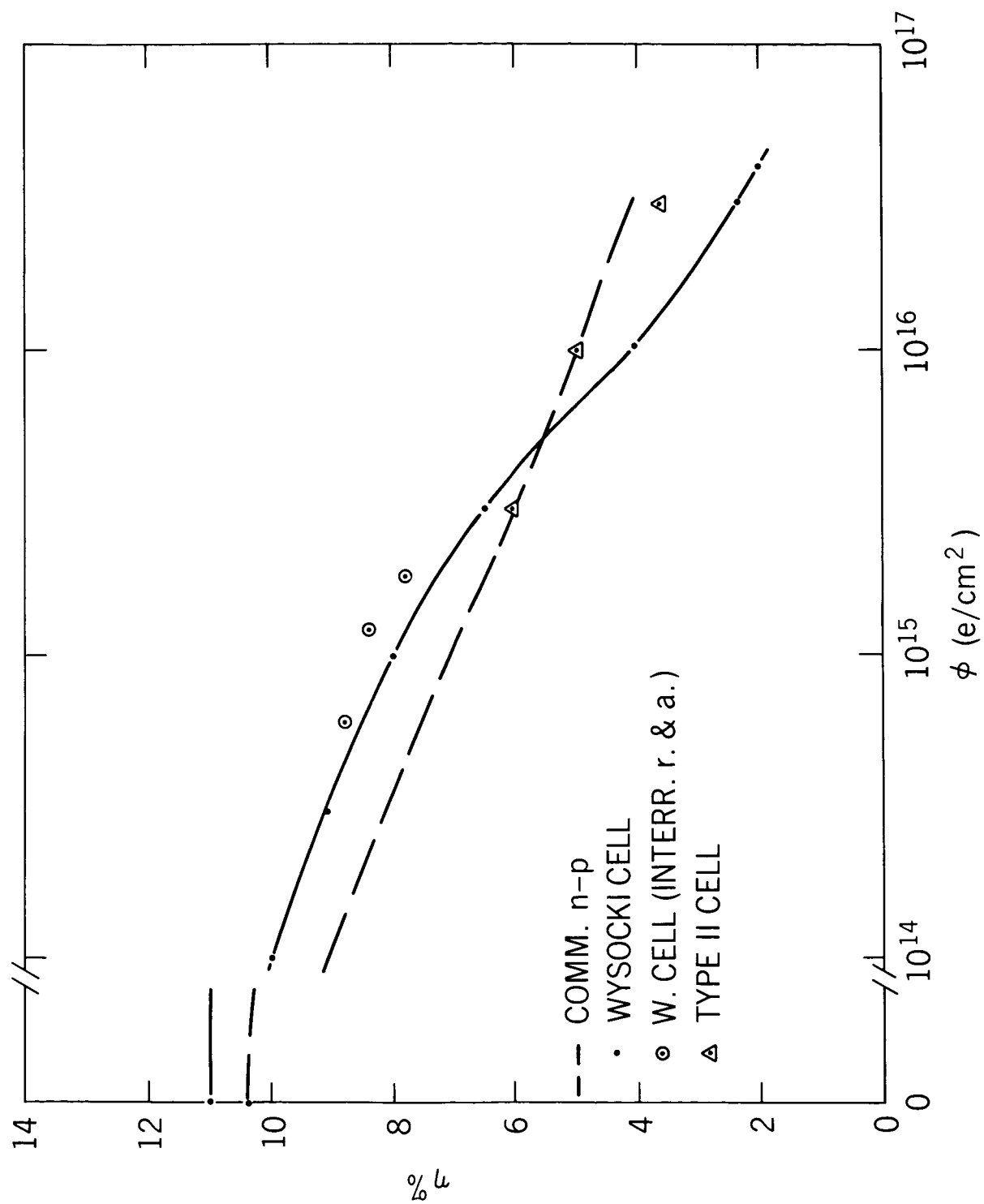
XI-4



XI-5



XI-6



XII. OUTLOOK

Some background material and our present knowledge have been discussed so far. We now face the inevitable question of the comparison between the merits of the Li cell and those of the current n-on-p cell. In order to discuss this rather premature problem, some points to be kept in mind are enumerated as follows.

1. Our knowledge of the structure of the defects as well as the mechanism of annealing is generally confined to some phenomenological observations. Therefore, we can predict neither the limit of the optimum efficiency in the initial performance nor the extent of the recovery which can be expected. This gap will be filled by an extensive program of basic studies centered on the following topics: Absorption band structure, local mode calculation, electron spin resonance, and Hall measurement of interstitial or displaced Li paired with radiation-induced defects in the same Si. Some of these programs are in progress and others are in the active planning stage.

2. More quantitative measurements on solar cells will be made, with special emphasis on stability versus precipitation with regard to Li or pairing of Li with other defects. The flux dependence of the stability, especially at a very high flux level, will be a most important problem.

3. Because of the observation of the dependence of the introduction rate as well as of the annealing on the defect concentration, analysis of the radiation damage requires a very specific description of the damage introduction rate and annealing condition. This includes the level of photon injection which produces a photovoltaic biased voltage; this in turn will influence the diffusion and the distribution of Li.

4. Radiation damage due to different types of charged particle is interesting especially in the case of high-rate irradiation, as in the encounter with a solar flare, or in the case of neutron damage, where the defect density in some microscopic region could exceed that of Li.

In view of the above problems, it seems a correct conclusion that the important question of actual application of the Li solar cell in space should be held in abeyance for a period of 6 months to 1 year until results are received from the extensive program which is now well in progress.

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